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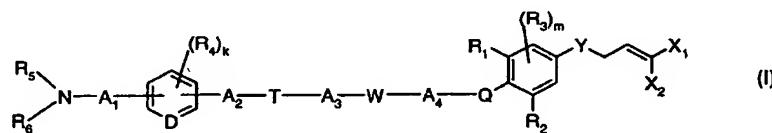
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(54) Title: 4-(3,3-DIHALO-ALLYLOXY)PHENOXY ALKYL DERIVATIVES



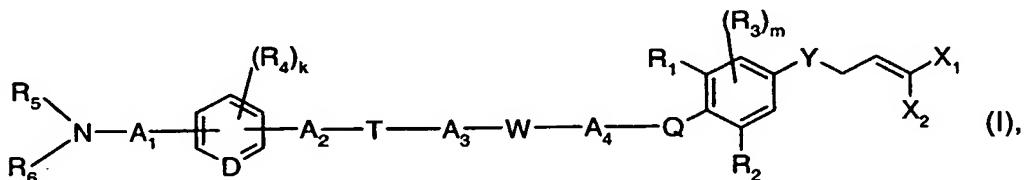
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(57) Abstract: Compounds of formula (I), wherein A<sub>1</sub>, A<sub>2</sub> and A<sub>3</sub> are each independently of the others a bond or a C<sub>1</sub>-C<sub>6</sub>alkylene bridge; A<sub>4</sub> is a C<sub>1</sub>-C<sub>6</sub>alkylene bridge; D is CH or N; W is, for example, O, NR<sub>7</sub> or S; T is, for example, a bond, O, NH or NR<sub>7</sub>; Q is O, NR<sub>7</sub>, S, SO or SO<sub>2</sub>; Y is O, NR<sub>7</sub>, S, SO, or SO<sub>2</sub>; X<sub>1</sub> and X<sub>2</sub> are each independently of the other fluorine, chlorine or bromine; R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> ar, for example, H, halogen, CN, nitro, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl or C<sub>2</sub>-C<sub>6</sub>alkenyl; R<sub>4</sub> is, for example, H, halogen, CN, nitro or C<sub>1</sub>-C<sub>6</sub>alkyl; R<sub>5</sub> and R<sub>6</sub> are, for example, H, CN, OH, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy or C<sub>1</sub>-C<sub>6</sub>haloalkoxy; R<sub>7</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl or C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl; k, when D is nitrogen, is 1, 2 or 3; or, when D is CH, is 1, 2, 3 or 4; and m is 1 or 2; and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form, a process for the preparation of those compounds and their use, pesticidal compositions in which the active ingredient has been selected from those compounds or an agrochemically acceptable salt thereof, a process for the preparation of those compositions and their use, plant propagation material treated with those compositions, and a method of controlling pests.

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4-(3,3-Dihalo-allyloxy)phenoxy Alkyl Derivatives

The present invention relates (1) to compounds of formula



wherein

$A_1$ ,  $A_2$  and  $A_3$  are each independently of the others a bond or a  $C_1$ - $C_6$ alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from  $C_3$ - $C_8$ cycloalkyl,  $C_3$ - $C_8$ cycloalkyl- $C_1$ - $C_6$ alkyl and  $C_1$ - $C_3$ haloalkyl;

$A_4$  is a  $C_1$ - $C_6$ alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from  $C_3$ - $C_8$ cycloalkyl,  $C_3$ - $C_8$ cycloalkyl- $C_1$ - $C_6$ alkyl and  $C_1$ - $C_3$ haloalkyl;

$D$  is CH or N;

$T$  is a bond, O, NH,  $NR_7$ , S, SO,  $SO_2$ ,  $-C(=O)-O-$ ,  $-O-C(=O)-$ ,  $-C(=O)-NR_8-$  or  $-NR_8-C(=O)-$ ;

$W$  is O,  $NR_7$ , S, SO,  $SO_2$ ,  $-C(=O)-O-$ ,  $-O-C(=O)-$ ,  $-C(=O)-NR_8-$  or  $-NR_8-C(=O)-$ ;

$Q$  is O,  $NR_7$ , S, SO or  $SO_2$ ;

$Y$  is O,  $NR_7$ , S, SO or  $SO_2$ ;

$X_1$  and  $X_2$  are each independently of the other fluorine, chlorine or bromine;

$R_1$ ,  $R_2$  and  $R_3$  are each independently of the others H, halogen, CN, nitro,  $C_1$ - $C_6$ alkyl,  $C_1$ - $C_6$ haloalkyl,  $C_1$ - $C_6$ alkylcarbonyl,  $C_2$ - $C_6$ alkenyl,  $C_2$ - $C_6$ haloalkenyl,  $C_2$ - $C_6$ alkynyl,  $C_1$ - $C_6$ alkoxy,  $C_1$ - $C_6$ haloalkoxy,  $C_2$ - $C_6$ alkenyloxy,  $C_2$ - $C_6$ haloalkenyloxy,  $C_2$ - $C_6$ alkynyloxy,  $C_1$ - $C_6$ alkoxycarbonyl or  $C_2$ - $C_6$ haloalkenyloxy; the substituents  $R_3$  being independent of one another when  $m$  is 2;

$R_4$  is H, halogen, CN, nitro,  $C_1$ - $C_6$ alkyl,  $C_1$ - $C_6$ haloalkyl,  $C_1$ - $C_6$ alkylcarbonyl,  $C_2$ - $C_6$ alkenyl,  $C_2$ - $C_6$ haloalkenyl,  $C_2$ - $C_6$ alkynyl,  $C_1$ - $C_6$ alkoxy,  $C_1$ - $C_6$ haloalkoxy,  $C_2$ - $C_6$ alkenyloxy,  $C_2$ - $C_6$ haloalkenyloxy,  $C_1$ - $C_6$ alkoxycarbonyl or  $C_2$ - $C_6$ haloalkenyloxy; the substituents  $R_4$  being independent of one another when  $k$  is greater than 1;

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R<sub>5</sub> is H, CN, OH, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyloxy, -C(=O)R<sub>9</sub>, -C(=S)R<sub>9</sub>, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group consisting of halogen, C<sub>1</sub>-C<sub>6</sub>alkyl, halo-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, halo-C<sub>1</sub>-C<sub>6</sub>alkoxy, hydroxy, cyano and nitro;

R<sub>6</sub> is H, CN, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, -C(=O)R<sub>9</sub>, -C(=S)R<sub>9</sub>, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group consisting of halogen, C<sub>1</sub>-C<sub>6</sub>alkyl, halo-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, halo-C<sub>1</sub>-C<sub>6</sub>alkoxy, hydroxy, cyano and nitro; or

R<sub>5</sub> and R<sub>6</sub> together form a four- to eight-membered alkylene or a four- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>, and the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl; or

R<sub>6</sub> is -C(=O)R<sub>9</sub> or -C(=S)R<sub>9</sub>, and R<sub>5</sub> and R<sub>9</sub> together form a two- to eight-membered alkylene or a two- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>, and wherein the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl; or

R<sub>5</sub> and R<sub>6</sub> are each independently of the other -C(=O)R<sub>9</sub> or -C(=S)R<sub>9</sub>, and the two R<sub>9</sub> together form a two- to eight-membered, straight-chain or branched alkylene or a two- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>; and wherein the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl;

R<sub>7</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl or C<sub>3</sub>-C<sub>8</sub>cycloalkyl;

R<sub>8</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl, -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl or C<sub>3</sub>-C<sub>8</sub>cycloalkyl;

R<sub>9</sub> is C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyloxy, C<sub>3</sub>-C<sub>6</sub>cycloalkyl, phenyl, benzyl; or phenyl or benzyl each of which is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>1</sub>-C<sub>6</sub>alkoxycarbonyl, C<sub>1</sub>-C<sub>3</sub>haloalkoxycarbonyl and C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy;

R<sub>10</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl or C<sub>3</sub>-C<sub>6</sub>cycloalkyl;

k, when D is nitrogen, is 1, 2 or 3; or, when D is CH, is 1, 2, 3 or 4; and

m is 1 or 2;

and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form, to a process for the preparation of those compounds, E/Z isomers and tautomers and to their use, to pesticidal compositions in which the active ingredient has been selected from those compounds, E/Z isomers and tautomers, and to a process for the preparation of those compositions and to their use, to intermediates and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in free form or in salt form, for the preparation of those compounds, where applicable to tautomers, in free form or in salt form, of those intermediates and to a process for the preparation of those intermediates and their tautomers and to their use.

Certain dihalovinyl derivatives are proposed in the literature as active ingredients in pesticidal compositions. The biological properties of those known compounds are not entirely satisfactory in the field of pest control, however, for which reason there is a need to provide further compounds having pesticidal properties, especially for controlling insects and members of the order Acarina, that problem being solved according to the invention by the provision of the present compounds of formula (I).

The compounds of formula (I) and, where applicable, their tautomers are able to form salts, e.g. acid addition salts. The latter are formed, for example, with strong inorganic acids, such as mineral acids, e.g. sulfuric acid, a phosphoric acid or a hydrohalic acid, with strong organic carboxylic acids, such as unsubstituted or substituted, e.g. halo-substituted, C<sub>1</sub>-C<sub>6</sub>alkanecarboxylic acids, for example acetic acid, saturated or unsaturated dicarboxylic acids, e.g. oxalic, malonic, maleic, fumaric or phthalic acid, hydroxycarboxylic acids, e.g.

ascorbic, lactic, malic, tartaric or citric acid, or benzoic acid, or with organic sulfonic acids, such as unsubstituted or substituted, e.g. halo-substituted, C<sub>1</sub>-C<sub>4</sub>alkane- or aryl-sulfonic acids, e.g. methane- or p-toluene-sulfonic acid. Furthermore, compounds of formula (I) having at least one acid group are able to form salts with bases. Suitable salts with bases are, for example, metal salts, such as alkali metal or alkaline earth metal salts, e.g. sodium, potassium or magnesium salts, or salts with ammonia or an organic amine, such as morpholine, piperidine, pyrrolidine, a mono-, di- or tri-lower alkylamine, e.g. ethyl-, diethyl-, triethyl- or dimethyl-propyl-amine, or a mono-, di- or tri-hydroxy-lower alkylamine, e.g. mono-, di- or tri-ethanolamine. It may also be possible for corresponding internal salts to be formed. The free form is preferred. Of the salts of compounds of formula (I), preference is given to agro-chemically advantageous salts. Hereinabove and hereinbelow any reference to the free compounds of formula (I) or to their salts is to be understood as including, where appropriate, the corresponding salts or the free compounds of formula (I), respectively. The same applies to tautomers of compounds of formula (I) and their salts.

The general terms used hereinabove and hereinbelow have the meanings given below, unless defined otherwise.

Halogen, as a group *per se* and as a structural element of other groups and compounds, such as haloalkyl, halocycloalkyl, haloalkenyl, haloalkynyl and haloalkoxy, is fluorine, chlorine, bromine or iodine, especially fluorine, chlorine or bromine, more especially fluorine or chlorine, especially chlorine.

Unless defined otherwise, carbon-containing groups and compounds each contain from 1 up to and including 20, preferably from 1 up to and including 18, especially from 1 up to and including 10, more especially from 1 up to and including 6, especially from 1 up to and including 4, especially from 1 up to and including 3, more especially 1 or 2, carbon atoms; methyl is especially preferred.

Alkyl, as a group *per se* and as a structural element of other groups and compounds, such as haloalkyl, alkoxy, alkoxyalkyl, haloalkoxy, alkoxycarbonyl, alkylthio, haloalkylthio, alkylsulfonyl and alkylsulfonyloxy, is - in each case giving due consideration to the number of carbon atoms contained in the group or compound in question - either straight-chained, e.g. methyl, ethyl, n-propyl, n-butyl, n-hexyl, n-octyl, n-decyl, n-dodecyl, n-hexadecyl or n-octadecyl, or branched, e.g. isopropyl, isobutyl, sec-butyl, tert-butyl, isopentyl, neopentyl or isohexyl.

Alkenyl and alkynyl - as groups *per se* and as structural elements of other groups and compounds, such as haloalkenyl, haloalkynyl, alkenyloxy, haloalkenyloxy, alkynyloxy or haloalkynyoxy - are straight-chain or branched and each contains two or preferably one unsaturated carbon-carbon bond(s). There may be mentioned by way of example vinyl, prop-2-en-1-yl, 2-methylprop-2-en-1-yl, but-2-en-1-yl, but-3-en-1-yl, prop-2-yn-1-yl, but-2-yn-1-yl and but-3-yn-1-yl.

Cycloalkyl - as a group *per se* and as a structural element of other groups and compounds, such as alkyl - is cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl or cyclooctyl. Cyclopentyl and cyclohexyl, and especially cyclopropyl, are preferred.

Alkylene is a straight-chain or branched bridging member and is especially -CH<sub>2</sub>-,-CH<sub>2</sub>CH<sub>2</sub>-,-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-,-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-,-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-,-CH(CH<sub>3</sub>)-,-CH(CH<sub>3</sub>)CH<sub>2</sub>-CH<sub>2</sub>-,-CH(C<sub>2</sub>H<sub>5</sub>)-,-C(CH<sub>3</sub>)<sub>2</sub>-,-CH(CH<sub>3</sub>)CH<sub>2</sub>-,-CH(CH<sub>3</sub>)CH(CH<sub>3</sub>)-, or -CH<sub>2</sub>C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>- Alkenylene is a straight-chain or branched bridging member and is for instance -CH=CH-, -CH=CH-CH<sub>2</sub>-,-CH<sub>2</sub>-CH=CH-CH<sub>2</sub>-,-CH<sub>2</sub>-CH=CH-CH-CH<sub>2</sub>-CH<sub>2</sub>-,-CH(CH<sub>3</sub>)CH=CH-.

Halo-substituted carbon-containing groups and compounds, such as haloalkyl and haloalkoxy, may be partially halogenated or perhalogenated, the halogen substituents in the case of polyhalogenation being the same or different. Examples of haloalkyl - as a group *per se* and as a structural element of other groups and compounds, such as haloalkoxy - are methyl substituted from one to three times by fluorine, chlorine and/or bromine, such as CHF<sub>2</sub>, CF<sub>3</sub> or CH<sub>2</sub>Cl; ethyl substituted from one to five times by fluorine, chlorine and/or bromine, such as CH<sub>2</sub>CF<sub>3</sub>, CF<sub>2</sub>CF<sub>3</sub>, CF<sub>2</sub>CCl<sub>3</sub>, CF<sub>2</sub>CHCl<sub>2</sub>, CF<sub>2</sub>CHF<sub>2</sub>, CF<sub>2</sub>CFCl<sub>2</sub>, CH<sub>2</sub>CH<sub>2</sub>Cl, CF<sub>2</sub>CHBr<sub>2</sub>, CF<sub>2</sub>CHClF, CF<sub>2</sub>CHBrF or CClFCHClF; propyl or isopropyl substituted from one to seven times by fluorine, chlorine and/or bromine, such as CH<sub>2</sub>CHBrCH<sub>2</sub>Br, CF<sub>2</sub>CHFCF<sub>3</sub>, CH<sub>2</sub>CF<sub>2</sub>CF<sub>3</sub>, CF<sub>2</sub>CF<sub>2</sub>CF<sub>3</sub>, CH(CF<sub>3</sub>)<sub>2</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>Cl; and butyl or an isomer thereof substituted from one to nine times by fluorine, chlorine and/or bromine, such as CF(CF<sub>3</sub>)CHFCF<sub>3</sub>, CF<sub>2</sub>(CF<sub>2</sub>)<sub>2</sub>CF<sub>3</sub> or CH<sub>2</sub>(CF<sub>2</sub>)<sub>2</sub>CF<sub>3</sub>.

Aryl is especially phenyl or naphthyl, preferably phenyl.

Preferred embodiments within the scope of the invention are

(2) compounds according to (1) of formula (I) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine or bromine, especially chlorine;

(3) compounds according to (1) or (2) of formula (I) wherein A<sub>1</sub> is a bond;

(4) compounds according to (1) or (2) of formula (I) wherein A<sub>1</sub> is -CH<sub>2</sub>CH<sub>2</sub>- or -CH<sub>2</sub>-, especially -CH<sub>2</sub>-;

(5) compounds according to (1) to (4) of formula (I) wherein the group A<sub>2</sub>-T-A<sub>3</sub> is a bond;

(6) compounds according to (1) to (5) of formula (I) wherein W is oxygen, -C(=O)O- or -C(=O)NH-, especially O;

(7) compounds according to (1) to (6) of formula (I) wherein A<sub>4</sub> is a straight-chain alkylene bridge, especially ethylene, propylene or butylene, more especially propylene;

(8) compounds according to (1) to (7) of formula (I) wherein Q is oxygen;

(9) compounds according to (1) to (8) of formula (I) wherein Y is oxygen;

(10) compounds according to (1) to (9) of formula (I) wherein R<sub>1</sub> and R<sub>2</sub> are bromine or chlorine, especially chlorine;

(11) compounds according to (1) to (10) of formula (I) wherein R<sub>3</sub> is hydrogen;

(12) compounds according to (1) to (11) of formula (I) wherein R<sub>4</sub> is hydrogen;

(13) compounds according to (1) to (12) of formula (I) wherein R<sub>5</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyloxy;

(14) compounds according to (1) to (13) of formula (I) wherein R<sub>6</sub> is -C(=O)R<sub>9</sub> or -C(=S)R<sub>9</sub>, and R<sub>9</sub> is C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyloxy, C<sub>3</sub>-C<sub>6</sub>cycloalkyl or phenyl;

(15) compounds according to (1) to (4) and (6) to (14) of formula (I) wherein A<sub>2</sub> is a bond, T is oxygen and A<sub>3</sub> is a C<sub>1</sub>-C<sub>6</sub>alkylene bridge;

(16) compounds according to (1) to (4) and (6) to (15) of formula (I) wherein A<sub>2</sub> is a bond, T is -C(=O)O- wherein the oxygen is bonded to A<sub>3</sub>, or is -C(=O)NH- wherein NH is bonded to A<sub>3</sub>, and A<sub>3</sub> is a C<sub>1</sub>-C<sub>6</sub>alkylene bridge;

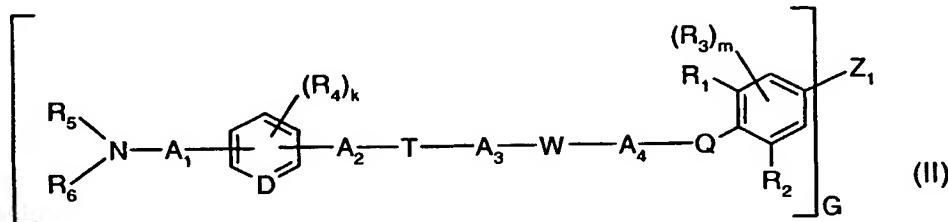
(17) compounds according to (1) to (16) of formula (I) wherein D is CH;

(18) compounds according to (1) to (16) of formula (I) wherein D is N.

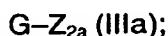
Special preference is given to the compounds listed in the Tables.

The invention relates also to a process for the preparation of a compound of formula (I), or a salt thereof, wherein

(a) a compound of formula

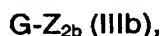


wherein A<sub>1</sub>, A<sub>2</sub>, A<sub>3</sub>, A<sub>4</sub>, D, W, Q, T, R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub>, R<sub>6</sub>, m and k are as defined for formula (I) under (1), Z<sub>1</sub> is  $-C(=O)R_{11}$  and R<sub>11</sub> is H or C<sub>1</sub>-C<sub>6</sub>alkyl, is converted in the presence of an oxidising agent, especially a peracid, into a compound of formula

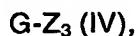


wherein Z<sub>2a</sub> is O-C(=O)-R<sub>12</sub> and R<sub>12</sub> is C<sub>1</sub>-C<sub>6</sub>alkyl, and G denotes the part of the formula in the brackets designated G in formula (II); either

(b<sub>1</sub>) a compound of formula (IIIa) above or of formula



wherein G denotes the part of the formula in the brackets designated G in formula (II), Z<sub>2b</sub> is a radical of formula  $-Y-C(=O)R_{13}$ , W is as defined for formula (I) under (1), and R<sub>13</sub> is C<sub>1</sub>-C<sub>12</sub>alkyl unsubstituted or substituted by from one to three identical or different halogen substituents, or is phenyl unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>1</sub>-C<sub>6</sub>alkoxycarbonyl and C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, is converted by hydrolytic cleavage into a compound of formula



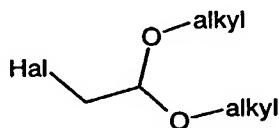
wherein G denotes the part of the formula in the brackets designated G in formula (II), Z<sub>3</sub> is YH, and Y is as defined for formula (I) under (1); or

(c) a compound of formula



wherein  $Z_4$  is  $Y\text{-CH}_2\text{-phenyl}$ , wherein the phenyl radical is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro,  $C_1\text{-}C_6$ alkyl,  $C_1\text{-}C_6$ haloalkyl,  $C_1\text{-}C_6$ alkylcarbonyl,  $C_2\text{-}C_6$ alkenyl,  $C_2\text{-}C_6$ haloalkenyl,  $C_2\text{-}C_6$ alkynyl,  $C_1\text{-}C_6$ alkoxy,  $C_1\text{-}C_6$ haloalkoxy,  $C_1\text{-}C_6$ alkoxycarbonyl or  $C_2\text{-}C_6$ haloalkenyloxy, G denotes the part of the formula in the brackets designated G in formula (II), and Y is as defined for formula (I), is converted by removal of the benzyl group into a compound of formula (IV), as defined above;

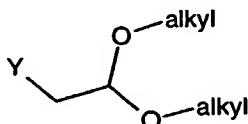
(d) the compound of formula (IV) so obtained is reacted in the presence of a base with a compound of formula



wherein Hal is halogen, preferably bromine or chlorine, and alkyl is  $C_1\text{-}C_6$ alkyl, or the two alkyl radicals together form a  $C_3\text{-}C_8$ alkylene bridge, to form a compound of formula

$G\text{-}Z_5$  (VI),

wherein G denotes the part of the formula in the brackets designated G in formula (II), and  $Z_5$  is



wherein alkyl and Y are as defined above;

(e) the compound of formula (VI) so obtained is converted by deprotection of the acetal function in the presence of an acid into a compound of formula

$G\text{-}Z_6$  (VII),

wherein  $Z_6$  is a group  $-Y\text{-CH}_2\text{-C}(=O)\text{H}$ , G is as defined above for the compound of formula (II), and Y is as defined for formula (I) under (1), either

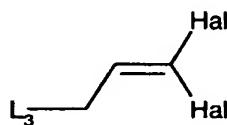
(f<sub>1</sub>) for the preparation of a compound of formula (I) wherein  $X_1$  and  $X_2$  are chlorine or bromine, a compound of formula (VII) is reacted in the presence of a phosphine with a compound of formula  $C(X)_4$  wherein X is chlorine or bromine; or

(f<sub>2</sub>) for the preparation of a compound of formula (I) wherein  $X_1$  and  $X_2$  are chlorine, a compound of formula (VII) is reacted first with  $CCl_3\text{-COOH}$  or with chloroform in the

presence of a strong base, then with acetic anhydride and subsequently with powdered zinc in acetic acid; or

(f<sub>3</sub>) for the preparation of a compound of formula (I) wherein X<sub>1</sub> is fluorine and X<sub>2</sub> is chlorine or bromine, a compound of formula (VII) is reacted first with a compound of the formula CF<sub>2</sub>X<sub>2</sub>, of the formula CFX<sub>3</sub>, of the formula CF<sub>2</sub>XC(=O)ONa or of the formula CFX<sub>2</sub>C(=O)ONa, in the presence of a phosphine; or

(g<sub>1</sub>) for the preparation of a compound of formula (I) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine or bromine, a compound of formula (IV) is reacted in the presence of base with a compound of formula



wherein L<sub>3</sub> is a leaving group, preferably chlorine or bromine, and Hal is chlorine or bromine; or

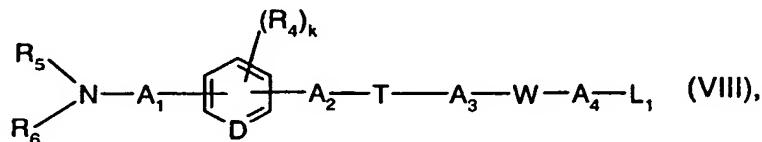
(g<sub>2</sub>) for the preparation of a compound of formula (I) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine or bromine, a compound of formula (IVa) or (IVb) is reacted in the presence of a base with a compound of formula



wherein Hal is halogen and X is chlorine or bromine.

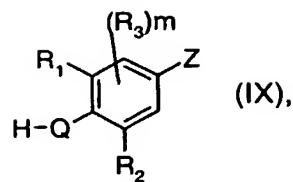
The invention relates also to

(h) a process for the preparation of a compound of formula (I), as defined under (1), and wherein Q is O, NR, or S, and R<sub>7</sub> is as defined for formula (I) under (1), wherein a compound of formula

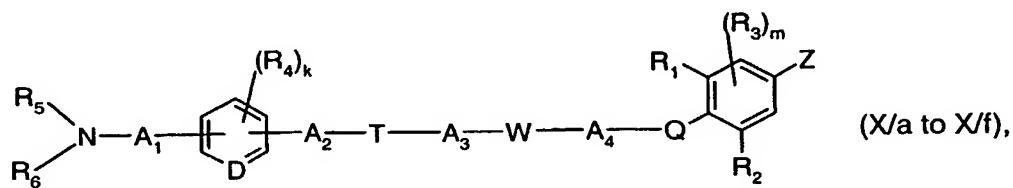


wherein A<sub>1</sub>, A<sub>2</sub>, A<sub>3</sub>, A<sub>4</sub>, D, W, T, R<sub>4</sub>, R<sub>5</sub>, R<sub>6</sub> and k are as defined for formula (I) under (1) and L<sub>1</sub> is a leaving group, is reacted in the presence of a base with a compound of formula

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wherein  $R_1$ ,  $R_2$ ,  $R_3$  and  $m$  are as defined for formula (I) under (1),  $Q$  is  $O$ ,  $NR_7$  or  $S$  and  $Z$  is one of the radicals  $Z_1$  to  $Z_6$  as defined for the above formulae (II) to (VII), and  $R_7$  is as defined for formula (I) under (1), and the resulting compound of formula

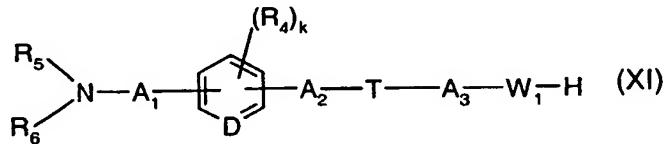


wherein  $A_1$ ,  $A_2$ ,  $A_3$ ,  $A_4$ ,  $D$ ,  $W$ ,  $Q$ ,  $T$ ,  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$ ,  $R_5$ ,  $R_6$ ,  $m$  and  $k$  are as defined for formula (I) under (1) and  $Z$  is one of the radicals  $Z_1$  to  $Z_6$  as defined for formulae (II) to (VII) indicated above, is, as necessary, that is to say according to the meaning of the radical  $Z$ , reacted further analogously to one or more of process steps (a) to (g).

In the compounds of formulae  $X/a$  to  $X/f$ ,  $Z$  in compound  $X/a$  has the same meanings as  $Z_1$  in the compound of formula (II), and  $Z$  in compound  $X/b$  has the same meanings as  $Z_2$  as defined for formula (III), and so on.

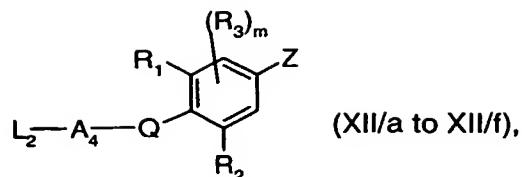
The invention relates also to

(i<sub>1</sub>) a process for the preparation of a compound of formula (I) as defined above wherein  $W$  is  $O$ ,  $NR_7$ ,  $S$ ,  $-O-C(=O)-$  or  $-NR_8-C(=O)-$ , and  $R_7$  and  $R_8$  are as defined for formula (I) under (1), wherein a compound of formula



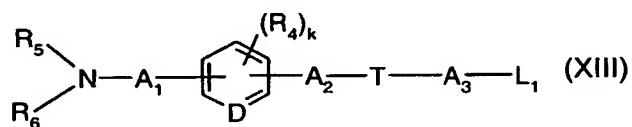
wherein  $A_1$ ,  $A_2$ ,  $A_3$ ,  $D$ ,  $T$ ,  $R_4$ ,  $R_5$ ,  $R_6$  and  $k$  are as defined for formula (I) under (1),  $W_1$  is  $O$ ,  $NR_7$ ,  $S$  or  $-NR_8-$  and  $R_7$  is as defined for formula (I) under (1), is reacted with a compound of formula

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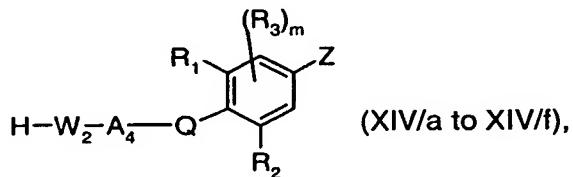


wherein  $A_4$ ,  $R_1$ ,  $R_2$ ,  $R_3$ ,  $Q$  and  $m$  are as defined for formula (I) under (1),  $L_2$  is a leaving group or a group  $\text{Hal}-\text{C}(=\text{O})-$  wherein  $\text{Hal}$  is a halogen atom, preferably chlorine or bromine, and  $Z$  is one of the radicals  $Z_1$  to  $Z_6$  as defined in formulae (II) to (VII) indicated above; or

(i<sub>2</sub>) for the preparation of a compound of formula (I) as defined above wherein  $W$  is O,  $\text{NR}_7$ , S,  $-\text{C}(=\text{O})-\text{O}-$  or  $-\text{C}(=\text{O})-\text{NR}_8-$ , and  $R_7$  and  $R_8$  are as defined for formula (I) under (1), wherein a compound of formula



wherein  $A_1$ ,  $A_2$ ,  $A_3$ ,  $D$ ,  $T$ ,  $R_4$ ,  $R_5$ ,  $R_6$  and  $k$  are as defined for formula (I) under (1), and  $L_1$  is a leaving group or a group  $-\text{C}(=\text{O})-\text{Hal}$  wherein  $\text{Hal}$  is a halogen atom, preferably chlorine or bromine, is reacted with a compound of formula



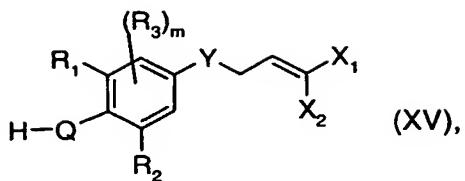
wherein  $W_2$  is O,  $\text{NR}_7$ , S or  $\text{NR}_8$ , and  $R_7$  and  $R_8$  are as defined for formula (I) under (1), and a resulting compound of formula (Xa) to (Xf) as defined above is, as necessary, that is to say according to the meaning of the radical  $Z$ , reacted further analogously to one or more of process steps (a) to (g).

In the compounds of formulae XII/a to XII/f and XIV/a to XIV/f, the radicals  $Z$  are as defined above for the compounds X/a to X/f; that is to say, for example,  $Z$  in the compound of formula XII/a has the same meanings as  $Z_1$  in the compound of formula (II), and  $Z$  in compound XII/b has the same meanings as  $Z_2$  as defined for formula (III), and so on.

The invention relates also to

(k) a process for the preparation of a compound of formula (I) as defined above under (1), wherein a compound of formula (VIII) as defined above is reacted in the presence of a base with a compound of formula

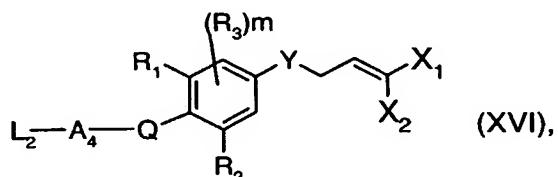
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wherein  $\text{R}_1$ ,  $\text{R}_2$ ,  $\text{R}_3$ ,  $\text{Q}$ ,  $\text{X}_1$ ,  $\text{X}_2$ ,  $\text{Y}$  and  $m$  are as defined for formula (I) under (1).

The invention also relates also to

(l) a process for the preparation of a compound of formula (I) as defined above under (1), wherein a compound of formula (XI) as defined above is reacted in a manner analogous to process variant (i) with a compound of formula



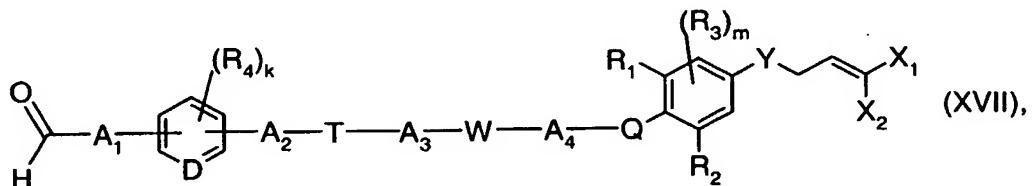
wherein  $\text{A}_4$ ,  $\text{R}_1$ ,  $\text{R}_2$ ,  $\text{R}_3$ ,  $\text{Q}$ ,  $\text{Y}$ ,  $\text{X}_1$ ,  $\text{X}_2$  and  $m$  are as defined for formula (I) under (1), and  $\text{L}_2$  is as defined for formula (XII).

The invention relates also to

(m) a process for the preparation of a compound of formulae (VIII), (X/a) bis X/f, (XI), (XIII), wherein  $\text{R}_5$  is  $-\text{C}(=\text{O})\text{R}_9$ , and  $\text{R}_6$  and  $\text{R}_9$  have the same meanings as given under (1) for formula (I), any of the compounds of the formulae (VIII), (X/a) to (X/f), (XI), (XIII), wherein  $\text{R}_5$  is H and  $\text{R}_6$  has the same meanings as given under (1) for formula (I), is reacted with an compound of the formula Halogen- $\text{C}(=\text{O})\text{R}_9$  or  $\text{O}(-\text{C}(=\text{O})\text{R}_9)_2$  in the presence of a base.

The invention relates also to

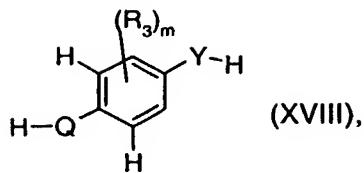
(n) a process for the preparation of a compound of formula (I), wherein  $\text{R}_6$  is hydrogen, wherein a compound of the formula



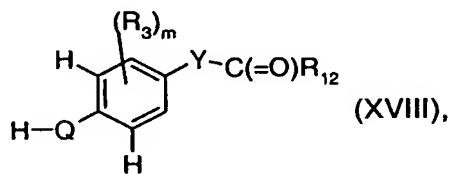
wherein  $\text{A}_1$ ,  $\text{A}_2$ ,  $\text{A}_3$ ,  $\text{A}_4$ ,  $\text{D}$ ,  $\text{W}$ ,  $\text{Q}$ ,  $\text{T}$ ,  $\text{R}_1$ ,  $\text{R}_2$ ,  $\text{R}_3$ ,  $\text{R}_4$ ,  $\text{X}_1$ ,  $\text{X}_2$ ,  $\text{Y}$ ,  $m$  and  $k$  are as defined for formula (I) under (1), is reacted with a primary amine in the presence of a reducing agent.

The compounds of formulae (IIIa) and (IIIb) wherein  $\text{R}_1$  and  $\text{R}_2$  are halogen can be obtained by reacting a compound of formula

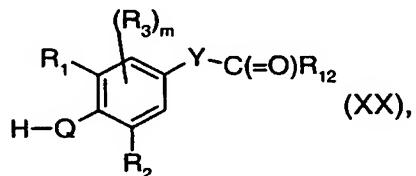
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wherein  $R_3$ ,  $Q$ ,  $Y$  and  $m$  are as defined for formula (I) under (1) with a compound of the formula  $\text{Hal}-\text{C}(=\text{O})-\text{R}_{12}$  wherein  $\text{R}_{12}$  is as defined above, halogenating the resulting compound of formula



wherein  $R_3$ ,  $Q$ ,  $Y$  and  $m$  are as defined for formula (I) under (1), and further reacting the resulting compound of formula



wherein  $R_3$ ,  $Q$ ,  $Y$  and  $m$  are as defined for formula (I) under (1) and  $R_1$  and  $R_2$  are halogen, analogously to Process (k).

The reactions described hereinabove and hereinbelow are carried out in a manner known *per se*, for example in the absence or, if necessary, in the presence of a suitable solvent or diluent or of a mixture thereof, the reactions being carried out, as required, with cooling, at room temperature or with heating, for example in a temperature range of approximately from  $-80^\circ\text{C}$  to the boiling temperature of the reaction mixture, preferably from approximately  $-20^\circ\text{C}$  to approximately  $+150^\circ\text{C}$ , and, if necessary, in a closed vessel, under pressure, under an inert gas atmosphere and/or under anhydrous conditions. Especially advantageous reaction conditions can be found in the Examples.

A leaving group, for example the leaving groups  $L_1$  and  $L_2$  defined above, or a counterion is to be understood hereinbefore and hereinbelow as being any removable group that customarily comes into consideration for chemical reactions, such as are known to the person skilled in the art; especially  $\text{OH}$ , halogens, such as fluorine, chlorine, bromine, iodine,

-O-Si(C<sub>1</sub>-C<sub>8</sub>alkyl)<sub>3</sub>, -O-aryl, -S-(C<sub>1</sub>-C<sub>8</sub>alkyl), -S-aryl, -O-S(=O)<sub>2</sub>U, -S(=O)U or -S(=O)<sub>2</sub>U, wherein U is unsubstituted or substituted C<sub>1</sub>-C<sub>8</sub>alkyl, C<sub>2</sub>-C<sub>8</sub>alkenyl, C<sub>2</sub>-C<sub>8</sub>alkynyl, unsubstituted or substituted aryl or unsubstituted or substituted benzyl. Especially preferred as leaving group are chlorine or bromine, mesylate, triflate, tosylate, especially chlorine; or chloride or bromide, especially chloride.

Process (a): The reaction is carried out in acetic acid or a halogenated hydrocarbon, such as dichloromethane, at temperatures of from -20°C to 100°C, preferably at from 20°C to 50°C. As oxidising agents there are used, for example, hydrogen peroxide, a peracid, such as peracetic acid, trifluoroperacetic acid, 3-chloroperbenzoic acid or a mixture thereof, such as sodium perborate in acetic acid.

Process (b): The reaction is preferably carried out in an alcohol, such as methanol, ethanol or an alcohol/water mixture, in the presence of an inorganic base, such as NaOH or KOH, and at temperatures of from 0°C to 150°C, preferably from 20°C to 80°C. Alternatively aminolysis with a primary amine, such as n-butylamine, can be carried out in a hydrocarbon, such as toluene or benzene, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C.

Process (c): Depending upon the nature of the benzyl substituent to be removed, the reaction can be carried out, for example, under a hydrogen atmosphere, at a pressure of from 1 to 150 bar, especially at from 1 to 20 bar, and with the addition of a catalyst, such as palladium/carbon, in an alcohol or ether. The preferred reaction temperature is from 0°C to 120°C, especially from 20°C to 80°C.

Processes (d) and (g): The reaction is preferably carried out in the presence of a base, such as potassium or sodium carbonate, in acetone or dimethylformamide, at temperatures of from 0°C to 150°C, preferably from 20°C to 80°C. If necessary, catalytic amounts of potassium iodide or sodium iodide, or phase transfer catalysts, such as crown ethers of quaternary ammonium salts, are added.

Process (e): The reaction is preferably carried out in acetone, dichloromethane, acetic acid, or especially in water, optionally with the addition of a mineral acid, at temperatures of from 0°C to 120°C, preferably at from 20°C to 50°C. For complete cleavage of the acetal it is preferable to add a strong mineral acid, for example hydrochloric acid, sulfuric acid or 4-toluenesulfonic acid.

Process (f): For the preparation of the difluoro-, dichloro-, dibromo-, chlorofluoro- and bromofluoro-vinyl compounds, reaction with  $\text{CCl}_4$ ,  $\text{CBr}_4$ ,  $\text{CF}_2\text{X}_2$ ,  $\text{CFX}_3$ ,  $\text{CF}_2\text{XC}(=\text{O})\text{ONa}$  or  $\text{CFX}_2\text{C}(=\text{O})\text{ONa}$  wherein X is bromine or chlorine is carried out in the presence of a trialkyl- or triaryl-phosphine, optionally with the addition of powdered zinc. The reaction is carried out in an inert solvent such as, for example, benzene or toluene, or an ether, such as diethyl ether, diisopropyl ether, dioxane or tetrahydrofuran, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C.

For the preparation of the dichlorovinyl compounds it is also possible for the process to be carried out in dimethylformamide, benzene, toluene, or in an ether, at temperatures of from 0°C to 120°C, preferably from 20°C to 80°C, and in the presence of trichloroacetic acid/ sodium trichloroacetate, then by addition of acetic anhydride, optionally with the addition of base, for example triethylamine, and finally by addition of zinc and acetic acid.

Processes (h) and (k): The reaction is preferably carried out in an ether, dimethylformamide, dimethylacetamide or N-methylpyrrolidone, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C, with the addition of a base, such as potassium or sodium carbonate. Alternatively a coupling reagent, for example azodicarboxylic acid diethyl or diisopropyl ester and triphenylphosphine, can be used.

Processes (i) and (l):

Where  $\text{L}_2$  is a group  $\text{Hal}-\text{C}(=\text{O})-$ , the process can be carried out in an inert solvent, such as in an ether or in toluene, at from 0°C to 80°C, and in the presence of a suitable base, for example a trialkylamine.

In the other cases the reaction is carried out in an ether, in an amide, such as dimethylformamide or N-methylpyrrolidone, and at from 0°C to 150°C. Sodium hydride, for example, can be used as base.

Process (m) and (n2): The reaction is preferably carried out in toluene, methylenchloride, chloroforme or in an ether at temperatures between 0°C and 120°C, preferably between 20°C and 80°C. Preferably a base such as triethylamine or sodiumcarbonate is added.

Process (n): The reaction is preferably carried out in tetrahydrofuran, dioxan, diethylether, toluene or in an alcoholes such as isopropanol, ethanol or methanol, at temperaturee between 0°C and 120°C, preferably between 20°C and 50°C. As reducing agent sodiumborhydride or hydrogen in the presence of a palladium catalyst can be used.

Compounds of formula (I) obtainable in accordance with the process or by other means can be converted into other compounds of formula (I) in a manner known *per se* by replacement of one or more substituents in the starting compound of formula (I) in customary manner by another (other) substituent(s) according to the invention.

In the case of such replacement, depending upon the choice of reaction conditions and starting materials suitable therefor, it is possible for only one substituent to be replaced by another substituent according to the invention in a reaction step or for a plurality of substituents to be replaced by other substituents according to the invention in the same reaction step.

Salts of compounds of formula (I) can be prepared in a manner known *per se*. For example, salts of compounds of formula (I) with bases are obtained by treatment of the free compounds with a suitable base or with a suitable ion exchange reagent.

Salts of compounds of formula (I) can be converted into the free compounds of formula (I) in customary manner, for example by treatment with a suitable acid or with a suitable ion exchange reagent.

Salts of compounds of formula (I) can be converted in a manner known *per se* into other salts of a compound of formula (I).

The compounds of formula (I), in free form or in salt form, may be in the form of one of the possible isomers or in the form of a mixture thereof, for example, depending on the number of asymmetric carbon atoms occurring in the molecule and their absolute and relative configuration, and/or depending on the configuration of non-aromatic double bonds occurring in the molecule, in the form of pure isomers, such as antipodes and/or diastereoisomers, or in the form of mixtures of isomers, such as mixtures of enantiomers, for example racemates, mixtures of diastereoisomers or mixtures of racemates. The invention relates both to the pure isomers and to all possible mixtures of isomers and is to be interpreted as such hereinbefore and hereinafter, even if stereochemical details are not mentioned specifically in every case.

Mixtures of diastereoisomers, mixtures of racemates and mixtures of double bond isomers of compounds of formula (I), in free form or in salt form, which may be obtained by the process according to the invention – depending upon the starting materials and procedures chosen – or by some other method, can be separated into the pure diastereoisomers or racemates in known manner on the basis of the physico-chemical differences

between the constituents, for example by means of fractional crystallisation, distillation and/or chromatography.

Mixtures of enantiomers, such as racemates, that are obtainable in a corresponding manner can be resolved into the enantiomers by known methods, for example by recrystallisation from an optically active solvent, by chromatography on chiral adsorbents, for example high pressure liquid chromatography (HPLC) on acetylcellulose, with the aid of suitable microorganisms, by cleavage with specific, immobilised enzymes, *via* the formation of inclusion compounds, for example using chiral crown ethers, only one enantiomer being complexed, or by conversion into diastereoisomeric salts and separation of the mixture of diastereoisomers so obtained, for example on the basis of their different solubilities by fractional crystallisation, into the diastereoisomers, from which the desired enantiomer can be freed by the action of suitable agents.

Apart from by separation of corresponding mixtures of isomers, pure diastereoisomers or enantiomers can be obtained according to the invention also by generally known methods of diastereoselective or enantioselective synthesis, for example by carrying out the process according to the invention using starting materials having correspondingly suitable stereochemistry.

In each case it is advantageous to isolate or synthesise the biologically more active isomer, e.g. enantiomer or diastereoisomer, or isomeric mixture, e.g. enantiomeric mixture or diastereoisomeric mixture, where the individual components have different biological activity.

The compounds of formula (I), in free form or salt form, can also be obtained in the form of their hydrates and/or may include other solvents, for example solvents which may have been used for the crystallisation of compounds in solid form.

The invention relates to all those embodiments of the process according to which a compound obtainable as starting material or intermediate at any stage of the process is used as starting material and some or all of the remaining steps are carried out or a starting material is used in the form of a derivative or salt and/or its racemates or antipodes or, especially, is formed under the reaction conditions.

In the process of the present invention it is preferable to use those starting materials and intermediates, in each case in free form or in salt form, which result in the compounds of formula (I) and their salts described at the beginning as being especially valuable.

The invention relates especially to the preparation processes described in Examples P1 to P4.

The invention relates also to the intermediates of formulae (II) to (XX), where novel, and, where applicable, to their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form. The preferences applying to such compounds are the same as those for the compounds of formula (I).

In the area of pest control, the compounds of formula (I) according to the invention are active ingredients exhibiting valuable preventive and/or curative activity with a very advantageous biocidal spectrum and a very broad spectrum, even at low rates of concentration, while being well tolerated by warm-blooded animals, fish and plants. They are, surprisingly, equally suitable for controlling both plant pests and ecto- and endo-parasites in humans and more especially in productive livestock, domestic animals and pets. They are effective against all or individual development stages of normally sensitive animal pests, but also of resistant animal pests, such as insects and representatives of the order Acarina, nematodes, cestodes and trematodes, while at the same time protecting useful organisms. The insecticidal or acaricidal activity of the active ingredients according to the invention may manifest itself directly, i.e. in the mortality of the pests, which occurs immediately or only after some time, for example during moulting, or indirectly, for example in reduced oviposition and/or hatching rate, good activity corresponding to a mortality of at least 50 to 60 %.

Successful control within the scope of the subject of the invention is possible, in particular, of pests from the orders Lepidoptera, Coleoptera, Orthoptera, Isoptera, Psocoptera, Anoplura, Mallophaga, Thysanoptera, Heteroptera, Homoptera, Hymenoptera, Diptera, Siphonaptera, Thysanura and Acarina, mainly Lepidoptera and Coleoptera. Very especially good control is possible of the following pests:

Abagrotis spp., Abraxas spp., Acantholeucania spp., Acanthoplusia spp., Acarus spp., Acarus siro, Aceria spp., Aceria sheldoni, Acleris spp., Acoloithus spp., Acompsia spp., Acossus spp., Acria spp., Acrobasis spp., Acrocercops spp., Acrolepia spp., Acrolepiopsis spp., Acronicta spp., Acropolitis spp., Actebia spp., Aculus spp., Aculus schlechtendali, Adoxophyes spp., Adoxophyes reticulana, Aedes spp., Aegeria spp., Aethes spp., Agapeta spp., Agonopterix spp., Agriopis spp., Agriotes spp., Agriphila spp., Agrochola spp., Agroperina spp., Alabama spp., Alabama argillaceae, Agrotis spp., Albuna spp., Alcathoe spp., Alcis spp., Aleimma spp., Aletia spp., Aleurothrixus spp., Aleurothrixus floccosus, Aleyrodes spp., Aleyrodes brassicae, Allophyes spp., Alsophila spp., Amata spp., Amathes spp.,

Amblyomma spp., Amblyptilia spp., Ammoconia spp., Amorbia spp., Amphion spp., Amphipoea spp., Amphipyra spp., Amyelois spp., Anacamptodes spp., Anagrapha spp., Anarsia spp., Anatrychytis spp., Anavitrinella spp., Ancylis spp., Andropolia spp., Anhimella spp., Antheraea spp., Antherigona spp., Antherigona soccata, Anthonomus spp., Anthonomus grandis, Anticarsia spp., Anticarsia gemmatalis, Aonidiella spp., Apamea spp., Aphania spp., Aphelia spp., Aphididae, Aphis spp., Apotomis spp., Aproaerema spp., Archippus spp., Archips spp., Acromyrmex, Arctia spp., Argas spp., Argolamprotes spp., Argyresthia spp., Argyrogramma spp., Argyroploce spp., Argyrotaenia spp., Arotrophora spp., Ascotis spp., Aspidiotus spp., Aspilapteryx spp., Asthenoptyla spp., Aterpia spp., Athetis spp., Atomaria spp., Atomaria linearis, Atta spp., Atypha spp., Autographa spp., Axylia spp., Bactra spp., Barbara spp., Batrachedra spp., Battaristis spp., Bembecia spp., Bernisia spp., Bernisia tabaci, Bibio spp., Bibio hortulanis, Bisigna spp., Blastesthia spp., Blatta spp., Blatella spp., Blepharosis spp., Bleptina spp., Boarmia spp., Bombyx spp., Bomolocha spp., Boophilus spp., Brachmia spp., Bradina spp., Brevipalpus spp., Brithys spp., Bryobia spp., Bryobia praetiosa, Bryotropha spp., Bupalus spp., Busseola spp., Busseola fusca, Cabera spp., Cacoecimorpha spp., Cadra spp., Cadra cautella, Caenurgina spp., Calipitrimerus spp., Callierges spp., Callophora spp., Callophora erythrocephala, Calophasia spp., Caloptilia spp., Calybites spp., Capnoptycha spp., Capua spp., Caradrina spp., Caripeta spp., Carmenta spp., Carposina spp., Carposina nipponensis, Catamacta spp., Catelaphris spp., Catoptria spp., Caustoloma spp., Celaena spp., Celypha spp., Cenopis spp., Cephus spp., Ceramica spp., Cerapteryx spp., Ceratitis spp., Ceratophyllus spp., Ceroplasma spp., Chaetocnema spp., Chaetocnema tibialis, Chamaesphecia spp., Charaneca spp., Cheimophila spp., Chersotis spp., Chiasmia spp., Chilo spp., Chionodes spp., Chrioptes spp., Choristoneura spp., Chrysaspidea spp., Chrysodeixis spp., Chrysomya spp., Chrysomphalus spp., Chrysomphalus dictyospermi, Chrysomphalus aonidium, Chrysoteuchia spp., Ciliix spp., Cimex spp., Clysia spp., Clysia ambiguella, Clepsis spp., Cnaemidophorus spp., Cnaphalocrocis spp., Cnephacia spp., Coccus spp., Coccus hesperidum, Cochylis spp., Coleophora spp., Colotois spp., Commophila spp., Conistra spp., Conopomorpha spp., Corcyra spp., Cornutiplusia spp., Cosmia spp., Cosmopolites spp., Cosmopterix spp., Cossus spp., Costaeonvexa spp., Crambus spp., Creatonotos spp., Crocidolomia spp., Crocidolomia binotalis, Croesia spp., Crymodes spp., Cryptaspasma spp., Cryptoblabes spp., Cryptocala spp., Cryptophlebia spp., Cryptophlebia leucotreta, Cryptoptila spp., Ctenopseustis spp., Cucullia spp., Curculio spp., Culex spp., Cuterebra spp., Cydia spp., Cydia pomonella, Cymbalophora spp., Dactylethora spp., Dacus spp., Dadica spp., Damalinea spp., Dasychira spp., Deca-

darchis spp., Decodes spp., Deilephila spp., Deltodes spp., Dendrolimus spp., Depressaria spp., Dermestes spp., Dermanyssus spp., Dermanyssus gallinae, Diabrotica spp., Diachrysia spp., Diaphania spp., Diarsia spp., Diasemia spp., Diatraea spp., Diceratura spp., Dichomeris spp., Dichrocrocis spp., Dichrorampha spp., Dicycla spp., Dioryctria spp., Diparopsis spp., Diparopsis castanea, Dipleurina spp., Diprion spp., Diprionidae, Discestra spp., Distantiella spp., Distantiella theobroma, Ditula spp., Diurnea spp., Doratopteryx spp., Drepana spp., Drosophila spp., Drosophila melanogaster, Dysauxes spp., Dysdercus spp., Dysstroma spp., Eana spp., Earias spp., Ecclitica spp., Ecdytolopha spp., Ecpyrorrhoe spp., Ectomyelois spp., Eetropis spp., Egira spp., Elasmopalpus spp., Emmelia spp., mpoasca spp., Empyreuma spp., Enargia spp., Enarmonia spp., Endopiza spp., Endothenia spp., Endotricha spp., Eoreuma spp., Eotetranychus spp., Eotetranychus carpini, Epagoge spp., Epelis spp., Ephestia spp., Ephestiodes spp., Epiblema spp., Epiehoristodes spp., Epinotia spp., Epiphyas spp., Epiplema spp., Epipsestis spp., Epirrhoe spp., Episimus spp., Epitymbia spp., Eplachna spp., Erannis spp., Erastria spp., Eremnus spp., Ereunetis spp., Eriophyes spp., Eriosoma spp., Eriosoma lanigerum, Erythroneura spp., Estigmene spp., Ethmia spp., Etiella spp., Euagrotis spp., Eucosma spp., Euehlaena spp., Euelidia spp., Eueosma spp., Euchistus spp., Eucosmomorpha spp., Eudonia spp., Eufidonia spp., Euhyponomeutoides spp., Eulepitodes spp., Eulia spp., Eulithis spp., Eupithecia spp., Euplexia spp., Eupoecilia spp., Eupoecilia ambiguella, Euproctis spp., Eupsilia spp., Eurhodope spp., Eurois spp., Eurygaster spp., Eurythmia spp., Eustrotia spp., Euxoa spp., Euzophera spp., Evergestis spp., Evippe spp., Exarterma spp., Fannia spp., Faronta spp., Feltia spp., Filatima spp., Fishia spp., Frankliniella spp., Fumibotys spp., Gaesa spp., Gasgardia spp., Gastrophilus spp., Gelechia spp., Gilpinia spp., Gilpinia polytorna, Glossina spp., Glyphipterix spp., Glyphodes spp., Gnorimoschemini spp., Gonodonta spp., Gortyna spp., Gracillaria spp., Graphania spp., Grapholita spp., Grapholitha spp., Gravitarata spp., Gretchenia spp., Griselda spp., Gryllotalpa spp., Gynaephora spp., Gypsonoma spp., Hada spp., Haematomitus spp., Halisdota spp., Harpipteryx spp., Harrisina spp., Hedyta spp., Helicoverpa spp., Heliothis spp., Hellula spp., Helotropa spp., Hemaris spp., Hercinothrips spp., Herculia spp., Hermonassa spp., Heterogenea spp., Holomelina spp., Homadaula spp., Homoeosoma spp., Homoglaea spp., Homohadena spp., Homona spp., Homonopsis spp., Hoplocampa spp., Hoplodrina spp., Hoshinoa spp., Hxalomma spp., Hydraecia spp., Hydriomena spp., Hyles spp., Hyloicus spp., Hypagyrtis spp., Hypatima spp., Hyphantria spp., Hyphantria cunea, Hypocala spp., Hypocoena spp., Hypodema spp., Hypobosca spp., Hypsipyla spp., Hyssia spp., Hysterosia spp., Idaea spp., Idia spp., Ipimorpha spp., Isia spp.,

Isochorista spp., Isophrictis spp., Isopolia spp., Isotrias spp., Ixodes spp., Itame spp., Jodia spp., Jodis spp., Kawabea spp., Keiferia spp., Keiferia lycopersicella, Labdia spp., Laciniopolia spp., Lambdina spp., Lamprothritpa spp., Laodelphax spp., Lasius spp., Laspeyresia spp., Leptinotarsa spp., Leptinotarsa decemlineata, Leptocorisa spp., Leptostales spp., Lecanium spp., Lecanium comi, Lepidosaphes spp., Lepisma spp., Lepisma saccharina , Lesmone spp., Leucania spp., Leucinodes spp., Leucophaea spp., Leucophaea maderae, Leucoptera spp., Leucoptera scitella, Linognathus spp., Liposcelis spp., Lissorhoptrus spp., Lithacodia spp., Lithocolletis spp., Lithomoia spp., Lithophane spp., Lixodessa spp., Lobesia spp., Lobesia botrana, Lobophora spp., Locusta spp., Lomanaltes spp., Lomographa spp., Loxagrotis spp., Loxostege spp., Lucilia spp., Lymantria spp., Lymnaecia spp., Lyonetia spp., Lyriomyza spp., Macdonnoughia spp., Macrauzata spp., Macro noctua spp., Macrosiphus spp., Malacosoma spp., Maliarpha spp., Mamestra spp., Mamestra brassicae, Manduca spp., Manduca sexta, Marasmia spp., Margaritia spp., Matratinea spp., Matsumuraes spp., Melanagromyza spp., Melipotes spp., Melissopus spp., Melittia spp., Melolontha spp., Meristis spp., Meritastis spp., Merophyas spp., Mesapamea spp., Mesogona spp., Mesoleuca spp., Metanema spp., Metendothenia spp., Metzneria spp., Micardia spp., Microcorses spp., Microleon spp., Mnesictena spp., Mocis spp., Monima spp., Monochroa spp., Monomorium spp., Monomorium pharaonis, Monopsis spp., Morrisonia spp., Musca spp., Mutuuraia spp., Myelois spp., Mythimna spp., Myzus spp., Naranga spp., Nedra spp., Nemapogon spp., Neodiprion spp., Neosphaleroptera spp., Nephelodes spp., Nephrotettix spp., Nezara spp., Nilaparvata spp., Niphonympha spp., Nippoptilia spp., Noctua spp., Nola spp., Notocelia spp., Notodonta spp., Nudaurelia spp., Ochropleura spp., Ocnerostoma spp., Oestrus spp., Olethreutes spp., Oligia spp., Olindia spp., Olygonychus spp., Olygonychus gallinae, Oncocnemis spp., Operophtera spp., Ophisma spp., Opogona spp., Oraesia spp., Orniodoros spp., Orgyia spp., Oria spp., Orseolia spp., Orthodes spp., Orthogonia spp., Orthosia spp., Oryzaephilus spp., Oscinella spp., Oscinella frit, Osminia spp., Ostrinia spp., Ostrinia nubilalis, Otiorhynchus spp., Ourapteryx spp., Pachetra spp., Pachysphinx spp., Pagyda spp., Paleacrita spp., Paliga spp., Palthis spp., Pammene spp., Pandemis spp., Panemeria spp., Panolis spp., Panolis flammea, Panonychus spp., Parargyresthia spp., Paradiarsia spp., Paralobesia spp., Paranthrene spp., Parapandemis spp., Parapediasia spp., Parastichtis spp., Parasyndemis spp., Paratoria spp., Pareromerme spp., Pectinophora spp., Pectinophora gossypiella, Pediculus spp., Pegomyia spp., Pegomyia hyoscyami, Pelochrista spp., Pennisetia spp., Penstemonia spp., Pemphigus spp., Peribatodes spp., Peridroma spp., Perileucoptera spp., Periplaneta spp., Perizoma spp., Petrova spp., Pexi-

copia spp., Phalonia spp., Phalonidia spp., Phaneta spp., Phlyctaenia spp., Phlyctinus spp., Phorbia spp., Phragmatobia spp., Phricanthes spp., Phthorimaea spp., Phthorimaea operculella, Phylocnistis spp., Phyllocoptruta spp., Phyllocoptruta oleivora, Phyllonorycter spp., Phylophila spp., Phylloxera spp., Pieris spp., Pieris rapae, Piesma spp., Planococcus spp., Planotortrix spp., Platyedra spp., Platynota spp., Platyptilia spp., Platysenta spp., Plodia spp., Plusia spp., Plutella spp., Plutella xylostella, Pododesia spp., Polia spp., Popillia spp., Polymixis spp., Polyphagotarsonemus spp., Polyphagotarsonemus latus, Prays spp., Prionoxystus spp., Probola spp., Proceras spp., Prochoerodes spp., Proeulia spp., Proschistis spp., Proselenia spp., Proserpinus spp., Protagrotis spp., Proteoteras spp., Protobathra spp., Protoschinia spp., Pselaphorus spp., Pseudaletia spp., Pseudanthonomus spp., Pseudaternelia spp., Pseudaulacaspis spp., Pseudexentera spp., Pseudococcus spp., Pseudohermenias spp., Pseudoplusia spp., Psoroptes spp., Psylla spp., Psylliodes spp., Pterophorus spp., Ptycholoma spp., Pulvinaria spp., Pulvinaria aethiopica, Pyralis spp., Pyrausta spp., Pyrgotis spp., Pyrreferra spp., Pyrrharctia spp., Quadraspidiotus spp., Rancora spp., Raphia spp., Reticulitermes spp., Retinia spp., Rhagoletis spp., Rhagoletis pomonella, Rhipicephalus spp., Rhizoglyphus spp., Rhizopertha spp., Rhodnius spp., Rhopalosiphum spp., Rhopobota spp., Rhyacia spp., Rhyacionia spp., Rhynchopacha spp., Rhysthenes spp., Rivula spp., Rondotia spp., Rusidrina spp., Rynchaglaea spp., Sabulodes spp., Sahlbergella spp., Sahlbergella singularis, Saissetia spp., Samia spp., Sannina spp., Sanninoidea spp., Sapoideus spp., Sarcoptes spp., Sathrobrota spp., Scarabeidae, Scelioodes spp., Schinia spp., Schistocerca spp., Schizaphis spp., Schizura spp., Schreckensteinia spp., Sciara spp., Scirpophaga spp., Scirthrips auranti, Scoparia spp., Scopula spp., Scotia spp., Scotinophara spp., Scotogramma spp., Scrobipalpa spp., Scrobipalopsis spp., Semiothisa spp., Sereda spp., Sesamia spp., Sesia spp., Sicya spp., Sideridis spp., Simyra spp., Sineugrapha spp., Sitochroa spp., Sitobion spp., Sitophilus spp., Sitotroga spp., Solenopsis spp., Smerinthus spp., Sophronia spp., Spaelotis spp., Spargaloma spp., Sparganothis spp., Spatialistis spp., Sperchia spp., Sphecia spp., Sphinx spp., Spilonota spp., Spodoptera spp., Spodoptera littoralis, Stagmatophora spp., Staphylinochrous spp., Stathmopoda spp., Stenodes spp., Sterrhia spp., Stomoxys spp., Strophedra spp., Sunira spp., Sutyna spp., Swammerdamia spp., Syllomatia spp., Sympistis spp., Synanthonedon spp., Synaxis spp., Syncopacma spp., Syndemis spp., Sygrapha spp., Synthomeida spp., Tabanus spp., Taeniarchis spp., Taeniothrips spp., Tannia spp., Tarsonemus spp., Tegulifera spp., Tehama spp., Teleiodes spp., Telorta spp., Tenebrio spp., Tephritis spp., Teratoglaea spp., Terricula spp., Tethea spp., Tetranychus spp., Thalpophila spp., Thaumetopoea spp., Thiodia

spp., Thrips spp., Thrips palmi, Thrips tabaci, Thyridopteryx spp., Thyris spp., Tineola spp., Tipula spp., Tortricidia spp., Tortrix spp., Trachea spp., Trialeurodes spp., Trialeurodes vaporariorum, Triatoma spp., Triaxomera spp., Tribolium spp., Tricodectes spp., Trichoplusia spp., Trichoplusia ni, Trichoptilus spp., Trioza spp., Trioza erytreae, Triphaenia spp., Triphosa spp., Trogoderma spp., Tyria spp., Udea spp., Unaspis spp., Unaspis citri, Utetheisa spp., Valeriodes spp., Vespa spp., Vespamima spp., Vitacea spp., Vitula spp., Witlesia spp., Xanthia spp., Xanthorhoe spp., Xanthotype spp., Xenomicta spp., Xenopsylla spp., Xenopsylla cheopsis, Xestia spp., Xylena spp., Xylomyges spp., Xyrosaris spp., Yponomeuta spp., Ypsolopha spp., Zale spp., Zanclognathus spp., Zeiraphera spp., Zenodoxus spp., Zeuzera spp., Zygaena spp.,

It is also possible to control pests of the class Nematoda using the compounds according to the invention. Such pests include, for example,

root knot nematodes, cyst-forming nematodes and also stem and leaf nematodes; especially of Heterodera spp., e.g. Heterodera schachtii, Heterodora avenae and Heterodora trifolii; Globodera spp., e.g. Globodera rostochiensis; Meloidogyne spp., e.g. Meloidogyne incognita and Meloidogyne javanica; Radopholus spp., e.g. Radopholus similis; Pratylenchus, e.g. Pratylenchus neglectans and Pratylenchus penetrans; Tylenchulus, e.g. Tylenchulus semipenetrans; Longidorus, Trichodorus, Xiphinema, Ditylenchus, Apheenchoides and Anguina; especially Meloidogyne, e.g. Meloidogyne incognita, and Heterodera, e.g. Heterodera glycines.

An especially important aspect of the present invention is the use of the compounds of formula (I) according to the invention in the protection of plants against parasitic feeding pests.

The action of the compounds according to the invention and the compositions comprising them against animal pests can be significantly broadened and adapted to the given circumstances by the addition of other insecticides, acaricides or nematicides. Suitable additives include, for example, representatives of the following classes of active ingredient: organophosphorus compounds, nitrophenols and derivatives, formamidines, ureas, carbamates, pyrethroids, chlorinated hydrocarbons, neonicotinoids and *Bacillus thuringiensis* preparations.

Examples of especially suitable mixing partners include: azamethiphos; chlорfenvinphos; cypermethrin, cypermethrin high-cis; cyromazine; diafenthiuron; diazinon; dichlorvos;

dicrotophos; dicyclanil; fenoxy carb; fluazuron; furathiocarb; isazofos; iodfenphos; kinoprene; iufenuron; methacryphon; methidathion; monocrotophos; phosphamidon; profenofos; diofenolan; a compound obtainable from the *Bacillus thuringiensis* strain GC91 or from strain NCTC11821; pymetrozine; bromopropylate; methoprene; disulfoton; quinalphos; tau-fluvalinate; thiocyclam; thiometon; aldicarb; azinphos-methyl; benfuracarb; bifenthrin; buprofezin; carbofuran; dibutylaminothio; cartap; chlorfluazuron; chlorpyrifos; cyfluthrin; lambda-cyhalothrin; alpha-cypermethrin; zeta-cypermethrin; deltamethrin; diflubenzuron; endosulfan; ethofencarb; fenitrothion; fenobucarb; fenvalerate; formothion; methiocarb; heptenophos; imidacloprid; thiamethoxam; clothianidin; isoprocarb; methamidophos; methomyl; mevinphos; parathion; parathion-methyl; phosalone; pirimicarb; propoxur; teflubenzuron; terbufos; triazamate; fenobucarb; tebufenozone; fipronil; beta-cyfluthrin; silafluofen; fenpyroximate; pyridaben; fenazaquin; pyriproxyfen; pyrimidifen; nitenpyram; acetamiprid; emamectin; emamectin-benzoate; spinosad; a plant extract that is active against insects; a preparation that comprises nematodes and is active against insects; a preparation obtainable from *Bacillus subtilis*; a preparation that comprises fungi and is active against insects; a preparation that comprises viruses and is active against insects; chlorfenapyr; acephate; acrinathrin; alanycarb; alphamethrin; amitraz; AZ 60541; azinphos A; azinphos M; azocyclotin; bendiocarb; bensultap; beta-cyfluthrin; BPMC; brofenprox; bromophos A; bufencarb; butocarboxin; butylpyridaben; cadusafos; carbaryl; carbophenothion; chloethocarb; chlorethoxyfos; chlormephos; cis-resmethrin; clocythrin; clofentezine; cyanophos; cycloprothrin; cyhexatin; demeton M; demeton S; demeton-S-methyl; dichlofenthion; dicliphos; diethion; dimethoate; dimethylvinphos; dioxathion; edifenphos; esfenvalerate; ethion; ethofenprox; ethoprophos; etrimphos; fenamiphos; fenbutatin oxide; fenothiocarb; fenpropothrin; fenpyrad; fenthion; fluazinam; flucycloxuron; flucythrinate; flufenoxuron; flufenprox; fonophos; fosthiazate; fubfenprox; HCH; hexaflumuron; hexythiazox; IKI-220; iprobenfos; isofenphos; isoxathion; ivermectin; malathion; mecarbam; mesulfenphos; metaldehyde; metolcarb; milbemectin; moxidectin; naled; NC 184; omethoate; oxamyl; oxydemethon M; oxydeprofos; permethrin; phentoate; phorate; phosmet; phoxim; pirimiphos M; pirimiphos E; promecarb; propaphos; prothiofos; prothoate; pyrachlophos; pyradaphenthion; pyresmethrin; pyrethrum; tebufenozone; salithion; sebufos; sulfotep; sulprofos; tebufenpyrad; tebupirimphos; tefluthrin; temephos; terbam; tetrachlorvinphos; thiacloprid; thiafenox; thiadicarb; thifanox; thionazin; thuringiensin; tralomethrin; triarathene; triazophos; triazuron; trichlorfon; triflumuron; trimethacarb; vamidothion; xylylcarb; YI

5301/5302; zetamethrin; DPX-MP062 — indoxacarb; methoxyfenozide; bifenzuron; XMC (3,5-xylyl methylcarbamate); or the fungus pathogen *Metarhizium anisopliae*.

The compounds according to the invention can be used to control, i.e. to inhibit or destroy, pests of the mentioned type occurring on plants, especially on useful plants and ornamentals in agriculture, in horticulture and in forestry, or on parts of such plants, such as the fruits, blossoms, leaves, stems, tubers or roots, while in some cases plant parts that grow later are still protected against those pests.

Target crops include especially cereals, such as wheat, barley, rye, oats, rice, maize and sorghum; beet, such as sugar beet and fodder beet; fruit, e.g. pomes, stone fruit and soft fruit, such as apples, pears, plums, peaches, almonds, cherries and berries, e.g. strawberries, raspberries and blackberries; leguminous plants, such as beans, lentils, peas and soybeans; oil plants, such as rape, mustard, poppy, olives, sunflowers, coconut, castor oil, cocoa and groundnuts; cucurbitaceae, such as marrows, cucumbers and melons; fibre plants, such as cotton, flax, hemp and jute; citrus fruits, such as oranges, lemons, grapefruit and mandarins; vegetables, such as spinach, lettuce, asparagus, cabbages, carrots, onions, tomatoes, potatoes and paprika; lauraceae, such as avocado, cinnamon and camphor; and tobacco, nuts, coffee, aubergines, sugar cane, tea, pepper, vines, hops, bananas, natural rubber plants and ornamentals.

Further areas of use of the compounds according to the invention are the protection of stored goods and storerooms and the protection of raw materials, and also in the hygiene sector, especially the protection of domestic animals and productive livestock against pests of the mentioned type, more especially the protection of domestic animals, especially cats and dogs, from infestation by fleas, ticks and nematodes.

The invention therefore relates also to pesticidal compositions, such as emulsifiable concentrates, suspension concentrates, directly sprayable or dilutable solutions, spreadable pastes, dilute emulsions, wettable powders, soluble powders, dispersible powders, wettable powders, dusts, granules and encapsulations of polymer substances, that comprise at least one of the compounds according to the invention, the choice of formulation being made in accordance with the intended objectives and the prevailing circumstances.

The active ingredient is used in those compositions in pure form, a solid active ingredient, for example, in a specific particle size, or preferably together with at least one of the adjuvants customary in formulation technology, such as extenders, e.g. solvents or solid carriers, or surface-active compounds (surfactants). In the area of parasite control in

humans, domestic animals, productive livestock and pets it will be self-evident that only physiologically tolerable additives are used.

Solvents are, for example: non-hydrogenated or partly hydrogenated aromatic hydrocarbons, preferably fractions C<sub>8</sub> to C<sub>12</sub> of alkylbenzenes, such as xylene mixtures, alkylated naphthalenes or tetrahydronaphthalene, aliphatic or cycloaliphatic hydrocarbons, such as paraffins or cyclohexane, alcohols, such as ethanol, propanol or butanol, glycols and ethers and esters thereof, such as propylene glycol, dipropylene glycol ether, ethylene glycol or ethylene glycol monomethyl or -ethyl ether, ketones, such as cyclohexanone, isophorone or diacetone alcohol, strongly polar solvents, such as N-methylpyrrolid-2-one, dimethyl sulfoxide or N,N-dimethylformamide, water, non-epoxidized or epoxidized plant oils, such as non-epoxidized or epoxidized rapeseed, castor, coconut or soya oil, and silicone oils.

The solid carriers used, for example for dusts and dispersible powders, are as a rule natural rock powders, such as calcite, talc, kaolin, montmorillonite or attapulgite. Highly disperse silicic acids or highly disperse absorbent polymers can also be added to improve the physical properties. Granular adsorptive granule carriers are porous types, such as pumice, crushed brick, sepiolite or bentonite, and non-sorbent carrier materials are calcite or sand. A large number of granular materials of inorganic or organic nature can furthermore be used, in particular dolomite or comminuted plant residues.

Surface-active compounds are, depending on the nature of the active compound to be formulated, nonionic, cationic and/or anionic surfactants or surfactant mixtures with good emulsifying, dispersing and wetting properties. The surfactants listed below are to be regarded only as examples; many other surfactants which are customary in formulation technology and are suitable according to the invention are described in the relevant literature.

Nonionic surfactants are, in particular, polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, which can contain 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols. Substances which are furthermore suitable are water-soluble polyethylene oxide adducts, containing 20 to 250 ethylene glycol ether and 10 to 100 propylene glycol ether groups, on propylene glycol, ethylene diaminopolypropylene glycol and alkyl polypropylene glycol having 1 to 10 carbon atoms in the alkyl chain. The compounds mentioned usually contain 1 to 5 ethylene glycol units per propylene glycol unit. Examples are nonylphenol-polyethoxyethanols, castor oil

polyglycol ethers, polypropylene-polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol. Other substances are fatty acid esters of polyoxyethylene sorbitan, such as polyoxyethylene sorbitan trioleate.

The cationic surfactants are, in particular, quaternary ammonium salts which contain, as substituents, at least one alkyl radical having 8 to 22 C atoms and, as further substituents, lower, non-halogenated or halogenated alkyl, benzyl or lower hydroxyalkyl radicals. The salts are preferably in the form of halides, methyl-sulfates or ethyl-sulfates. Examples are stearyl-trimethyl-ammonium chloride and benzyl-di-(2-chloroethyl)-ethyl-ammonium bromide.

Suitable anionic surfactants can be both water-soluble soaps and water-soluble synthetic surface-active compounds. Suitable soaps are the alkali metal, alkaline earth metal and substituted or unsubstituted ammonium salts of higher fatty acids ( $C_{10}$ - $C_{22}$ ), such as the sodium or potassium salts of oleic or stearic acid, or of naturally occurring fatty acid mixtures, which can be obtained, for example, from coconut oil or tall oil; and furthermore also the fatty acid methyl-taurine salts. However, synthetic surfactants are more frequently used, in particular fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates. The fatty sulfonates and sulfates are as a rule in the form of alkali metal, alkaline earth metal or substituted or unsubstituted ammonium salts and in general have an alkyl radical of 8 to 22 C atoms, alkyl also including the alkyl moiety of acyl radicals; examples are the sodium or calcium salt of ligninsulfonic acid, of dodecylsulfuric acid ester or of a fatty alcohol sulfate mixture prepared from naturally occurring fatty acids. These also include the salts of sulfuric acid esters and sulfonic acids of fatty alcohol-ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonic acid groups and a fatty acid radical having about 8 to 22 C atoms. Alkylarylsulfonates are, for example, the sodium, calcium or triethanolammonium salts of dodecylbenzenesulfonic acid, of dibutylnaphthalenesulfonic acid or of a naphthalenesulfonic acid-formaldehyde condensation product. Corresponding phosphates, such as salts of the phosphoric acid ester of a p-nonylphenol-(4-14)-ethylene oxide adduct or phospholipids, can further also be used.

The compositions as a rule comprise 0.1 to 99 %, in particular 0.1 to 95 %, of active compound and 1 to 99.9 %, in particular 5 to 99.9 %, of - at least - one solid or liquid auxiliary, it being possible as a rule for 0 to 25 %, in particular 0.1 to 20 %, of the composition to be surfactants (% is in each case per cent by weight). While concentrated compositions are more preferred as commercial goods, the end user as a rule uses dilute

compositions which comprise considerably lower concentrations of active compound.

Preferred compositions are composed, in particular, as follows (% = per cent by weight):

Emulsifiable concentrates:

|                    |                                |
|--------------------|--------------------------------|
| active ingredient: | 1 to 90%, preferably 5 to 20%  |
| surfactant:        | 1 to 30%, preferably 10 to 20% |
| solvent:           | 5 to 98%, preferably 70 to 85% |

Dusts:

|                    |                                     |
|--------------------|-------------------------------------|
| active ingredient: | 0.1 to 10%, preferably 0.1 to 1%    |
| solid carrier:     | 99.9 to 90%, preferably 99.9 to 99% |

Suspension concentrates:

|                    |                                 |
|--------------------|---------------------------------|
| active ingredient: | 5 to 75%, preferably 10 to 50%  |
| water:             | 94 to 24%, preferably 88 to 30% |
| surfactant:        | 1 to 40%, preferably 2 to 30%   |

Wettable powders:

|                    |                                 |
|--------------------|---------------------------------|
| active ingredient: | 0.5 to 90%, preferably 1 to 80% |
| surfactant:        | 0.5 to 20%, preferably 1 to 15% |
| solid carrier:     | 5 to 99%, preferably 15 to 98%  |

Granules:

|                    |                                   |
|--------------------|-----------------------------------|
| active ingredient: | 0.5 to 30%, preferably 3 to 15%   |
| solid carrier:     | 99.5 to 70%, preferably 97 to 85% |

The compositions according to the invention may also comprise further solid or liquid adjuvants, such as stabilisers, e.g. vegetable oils or epoxidised vegetable oils (e.g. epoxidised coconut oil, rapeseed oil or soybean oil), antifoams, e.g. silicone oil, preservatives, viscosity regulators, binders and/or tackifiers as well as fertilisers or other active ingredients for obtaining special effects, e.g. acaricides, bactericides, fungicides, nematicides, molluscicides or selective herbicides.

The crop protection products according to the invention are prepared in known manner, in the absence of adjuvants, e.g. by grinding, sieving and/or compressing a solid active ingredient or mixture of active ingredients, for example to a certain particle size, and in the presence of at least one adjuvant, for example by intimately mixing and/or grinding the

active ingredient or mixture of active ingredients with the adjuvant(s). The invention relates likewise to those processes for the preparation of the compositions according to the invention and to the use of the compounds of formula (I) in the preparation of those compositions.

The invention relates also to the methods of application of the crop protection products, i.e. the methods of controlling pests of the mentioned type, such as spraying, atomising, dusting, coating, dressing, scattering or pouring, which are selected in accordance with the intended objectives and the prevailing circumstances, and to the use of the compositions for controlling pests of the mentioned type. Typical rates of concentration are from 0.1 to 1000 ppm, preferably from 0.1 to 500 ppm, of active ingredient. The rates of application per hectare are generally from 1 to 2000 g of active ingredient per hectare, especially from 10 to 1000 g/ha, preferably from 20 to 600 g/ha.

A preferred method of application in the area of crop protection is application to the foliage of the plants (foliar application), the frequency and the rate of application being dependent upon the risk of infestation by the pest in question. However, the active ingredient can also penetrate the plants through the roots (systemic action) when the locus of the plants is impregnated with a liquid formulation or when the active ingredient is incorporated in solid form into the locus of the plants, for example into the soil, e.g. in granular form (soil application). In the case of paddy rice crops, such granules may be applied in metered amounts to the flooded rice field.

The crop protection products according to the invention are also suitable for protecting plant propagation material, e.g. seed, such as fruits, tubers or grains, or plant cuttings, against animal pests. The propagation material can be treated with the composition before planting: seed, for example, can be dressed before being sown. The active ingredients according to the invention can also be applied to grains (coating), either by impregnating the seeds in a liquid formulation or by coating them with a solid formulation. The composition can also be applied to the planting site when the propagation material is being planted, for example to the seed furrow during sowing. The invention relates also to such methods of treating plant propagation material and to the plant propagation material so treated.

The following Examples serve to illustrate the invention. They do not limit the invention. Temperatures are given in degrees Celsius; mixing ratios of solvents are given in parts by volume.

### Preparation Examples

Example P1): Preparation of (4-{3-[2,6-dichloro-4-(3,3-dichloroallyloxy)-phenoxy]-propoxy}-phenyl)-carbamic acid tert-butyl ester

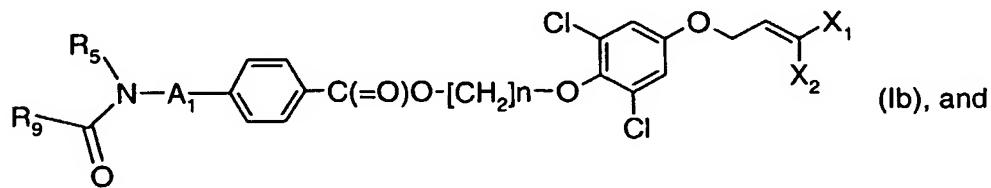
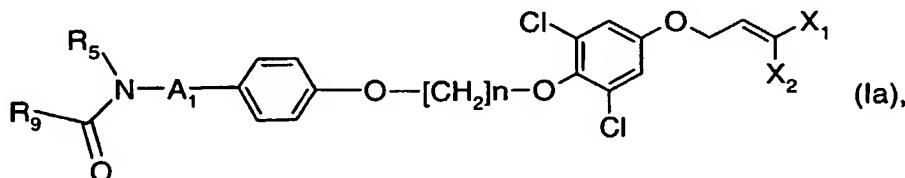
1.69 g of azadicarboxylic acid diisopropyl ester are added dropwise at 0-5°C to a solution of 1.97 g of triphenylphosphine in 60 ml of THF. After 30 minutes at 0-5°C, 2.5 g of 3-[2,6-dichloro-4-(3,3-dichloroallyloxy)-phenoxy]-propan-1-ol and 1.5 g of (4-hydroxy-phenyl)-carbamic acid tert-butyl ester dissolved in 30 ml of tetrahydrofuran are added dropwise. After being stirred for 24 hours at room temperature, the reaction mixture is concentrated and then purified over silica gel. (4-{3-[2,6-Dichloro-4-(3,3-dichloroallyloxy)-phenoxy]-propoxy}-phenyl)-carbamic acid tert-butyl ester (compound 1.6) is obtained.

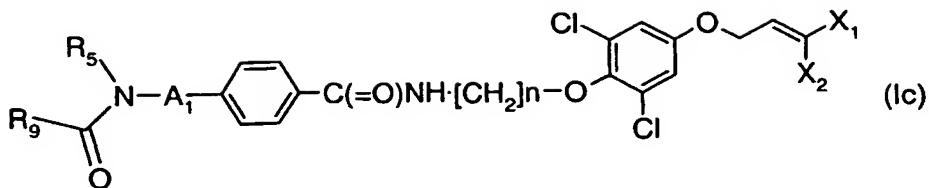
Preparation Example P2): Preparation of 3-(4-{3-[2,6-dichloro-4-(3,3-dichloroallyloxy)-phenoxy]-propoxy}-phenyl)-oxazolidin-2-one

7 mg of copper(I) iodide, 2 mg of ethylenediamine and 155 mg of tripotassium phosphate are added, under argon, to a solution of 200 mg of 1,3-dichloro-5-(3,3-dichloroallyloxy)-2-[3-(4-iodophenoxy)-propoxy]-benzene and 38 mg of 2-oxazolidinone in 4 ml of dioxane. After 17 hours at 110°C the reaction mixture is filtered and concentrated. Purification over silica gel yields the title compound (compound 29.3 ).

Example P3): In a manner analogous to that described above it is also possible to prepare the further compounds of Tables 2 to 28 below. In the tables,  $n_D^{20}$  is the refraction index and m.p. the melting point in °C.

Table A: Compounds of formula





| No.  | R <sub>9</sub>      | R <sub>5</sub> |
|------|---------------------|----------------|
| A.1  | H                   | H              |
| A.2  | methoxy             | H              |
| A.3  | ethoxy              | H              |
| A.4  | n-propoxy           | H              |
| A.5  | n-butoxy            | H              |
| A.6  | n-pentyloxy         | H              |
| A.7  | n-hexyloxy          | H              |
| A.8  | isopropoxy          | H              |
| A.9  | isobutoxy           | H              |
| A.10 | isopentyloxy        | H              |
| A.11 | tert-butoxy         | H              |
| A.12 | 2,2-dimethylpropoxy | H              |
| A.13 | vinyloxy            | H              |
| A.14 | allyloxy            | H              |
| A.15 | propargyloxy        | H              |
| A.16 | propen-2-yloxy      | H              |
| A.17 | benzyloxy           | H              |
| A.18 | 4-nitrobenzyloxy    | H              |
| A.19 | methyl              | H              |
| A.20 | ethyl               | H              |
| A.21 | n-propyl            | H              |
| A.22 | n-butyl             | H              |
| A.23 | n-pentyl            | H              |
| A.24 | n-hexyl             | H              |
| A.25 | isopropyl           | H              |
| A.26 | isobutyl            | H              |
| A.27 | isopentyl           | H              |
| A.28 | tert-butyl          | H              |
| A.29 | 2,2-dimethylpropyl  | H              |
| A.30 | cyclopropyl         | H              |
| A.31 | cyclobutyl          | H              |
| A.32 | cyclopentyl         | H              |
| A.33 | cyclohexyl          | H              |
| A.34 | trifluoromethyl     | H              |
| A.35 | pentafluoroethyl    | H              |

| No.  | R <sub>9</sub>                           | R <sub>5</sub> |
|------|--|----------------|
| A.36 | 4-trifluoromethylphenyl                  | H              |
| A.37 | 4-trifluoromethylbenzyl                  | H              |
| A.38 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | H              |
| A.39 | 2-chloro-1,1-dimethyl-ethyl              | H              |
| A.40 | 2-chlorophenyl                           | H              |
| A.41 | 2-fluorophenyl                           | H              |
| A.42 | 2,4-difluorophenyl                       | H              |
| A.43 | 4-chlorophenyl                           | H              |
| A.44 | 4-fluorophenyl                           | H              |
| A.45 | 2,4-dichlorophenyl                       | H              |
| A.46 | 4-trifluoromethoxyphenyl                 | H              |
| A.47 | H  | methyl         |
| A.48 | methoxy                                  | methyl         |
| A.49 | ethoxy                                   | methyl         |
| A.50 | n-propoxy                                | methyl         |
| A.51 | n-butoxy                                 | methyl         |
| A.52 | n-pentyloxy                              | methyl         |
| A.53 | n-hexyloxy                               | methyl         |
| A.54 | isopropoxy                               | methyl         |
| A.55 | isobutoxy                                | methyl         |
| A.56 | isopentyloxy                             | methyl         |
| A.57 | tert-butoxy                              | methyl         |
| A.58 | 2,2-dimethylpropoxy                      | methyl         |
| A.59 | vinyloxy                                 | methyl         |
| A.60 | allyloxy                                 | methyl         |
| A.61 | propargyloxy                             | methyl         |
| A.62 | propen-2-yloxy                           | methyl         |
| A.63 | benzyloxy                                | methyl         |
| A.64 | 4-nitrobenzyloxy                         | methyl         |
| A.65 | methyl                                   | methyl         |
| A.66 | ethyl                                    | methyl         |
| A.67 | n-propyl                                 | methyl         |
| A.68 | n-butyl                                  | methyl         |
| A.69 | n-pentyl                                 | methyl         |
| A.70 | n-hexyl                                  | methyl         |
| A.71 | isopropyl                                | methyl         |
| A.72 | isobutyl                                 | methyl         |
| A.73 | isopentyl                                | methyl         |
| A.74 | tert-butyl                               | methyl         |
| A.75 | 2,2-dimethylpropyl                       | methyl         |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.76  | cyclopropyl                              | methyl         |
| A.77  | cyclobutyl                               | methyl         |
| A.78  | cyclopentyl                              | methyl         |
| A.79  | cyclohexyl                               | methyl         |
| A.80  | trifluoromethyl                          | methyl         |
| A.81  | pentafluoroethyl                         | methyl         |
| A.82  | 4-trifluoromethylphenyl                  | methyl         |
| A.83  | 4-trifluoromethylbenzyl                  | methyl         |
| A.84  | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | methyl         |
| A.85  | 2-chloro-1,1-dimethyl-ethyl              | methyl         |
| A.86  | 2-chlorophenyl                           | methyl         |
| A.87  | 2-fluorophenyl                           | methyl         |
| A.88  | 2,4-difluorophenyl                       | methyl         |
| A.89  | 4-chlorophenyl                           | methyl         |
| A.90  | 4-fluorophenyl                           | methyl         |
| A.91  | 2,4-dichlorophenyl                       | methyl         |
| A.92  | 4-trifluoromethoxyphenyl                 | methyl         |
| A.93  | H  | ethyl          |
| A.94  | methoxy                                  | ethyl          |
| A.95  | ethoxy                                   | ethyl          |
| A.96  | n-propoxy                                | ethyl          |
| A.97  | n-butoxy                                 | ethyl          |
| A.98  | n-pentyloxy                              | ethyl          |
| A.99  | n-hexyloxy                               | ethyl          |
| A.100 | isopropoxy                               | ethyl          |
| A.101 | isobutoxy                                | ethyl          |
| A.102 | isopentyloxy                             | ethyl          |
| A.103 | tert-butoxy                              | ethyl          |
| A.104 | 2,2-dimethylpropoxy                      | ethyl          |
| A.105 | vinyloxy                                 | ethyl          |
| A.106 | allyloxy                                 | ethyl          |
| A.107 | propargyloxy                             | ethyl          |
| A.108 | propen-2-yloxy                           | ethyl          |
| A.109 | benzyloxy                                | ethyl          |
| A.110 | 4-nitrobenzyloxy                         | ethyl          |
| A.111 | methyl                                   | ethyl          |
| A.112 | ethyl                                    | ethyl          |
| A.113 | n-propyl                                 | ethyl          |
| A.114 | n-butyl                                  | ethyl          |
| A.115 | n-pentyl                                 | ethyl          |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.116 | n-hexyl                                  | ethyl          |
| A.117 | isopropyl                                | ethyl          |
| A.118 | isobutyl                                 | ethyl          |
| A.119 | isopentyl                                | ethyl          |
| A.120 | tert-butyl                               | ethyl          |
| A.121 | 2,2-dimethylpropyl                       | ethyl          |
| A.122 | cyclopropyl                              | ethyl          |
| A.123 | cyclobutyl                               | ethyl          |
| A.124 | cyclopentyl                              | ethyl          |
| A.125 | cyclohexyl                               | ethyl          |
| A.126 | trifluoromethyl                          | ethyl          |
| A.127 | pentafluoroethyl                         | ethyl          |
| A.128 | 4-trifluoromethylphenyl                  | ethyl          |
| A.129 | 4-trifluoromethylbenzyl                  | ethyl          |
| A.130 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | ethyl          |
| A.131 | 2-chloro-1,1-dimethyl-ethyl              | ethyl          |
| A.132 | 2-chlorophenyl                           | ethyl          |
| A.133 | 2-fluorophenyl                           | ethyl          |
| A.134 | 2,4-difluorophenyl                       | ethyl          |
| A.135 | 4-chlorophenyl                           | ethyl          |
| A.136 | 4-fluorophenyl                           | ethyl          |
| A.137 | 2,4-dichlorophenyl                       | ethyl          |
| A.138 | 4-trifluoromethoxyphenyl                 | ethyl          |
| A.139 | H  | propyl         |
| A.140 | methoxy                                  | propyl         |
| A.141 | ethoxy                                   | propyl         |
| A.142 | n-propoxy                                | propyl         |
| A.143 | n-butoxy                                 | propyl         |
| A.144 | n-pentyloxy                              | propyl         |
| A.145 | n-hexyloxy                               | propyl         |
| A.146 | Isopropoxy                               | propyl         |
| A.147 | Isobutoxy                                | propyl         |
| A.148 | isopentyloxy                             | propyl         |
| A.149 | tert-butoxy                              | propyl         |
| A.150 | 2,2-dimethylpropoxy                      | propyl         |
| A.151 | Vinyloxy                                 | propyl         |
| A.152 | Allyloxy                                 | propyl         |
| A.153 | propargyloxy                             | propyl         |
| A.154 | propen-2-yloxy                           | propyl         |
| A.155 | benzyloxy                                | propyl         |

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| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.156 | 4-nitrobenzyloxy                         | propyl         |
| A.157 | methyl                                   | propyl         |
| A.158 | ethyl                                    | propyl         |
| A.159 | n-propyl                                 | propyl         |
| A.160 | n-butyl                                  | propyl         |
| A.161 | n-pentyl                                 | propyl         |
| A.162 | n-hexyl                                  | propyl         |
| A.163 | isopropyl                                | propyl         |
| A.164 | isobutyl                                 | propyl         |
| A.165 | isopentyl                                | propyl         |
| A.166 | tert-butyl                               | propyl         |
| A.167 | 2,2-dimethylpropyl                       | propyl         |
| A.168 | cyclopropyl                              | propyl         |
| A.169 | cyclobutyl                               | propyl         |
| A.170 | cyclopentyl                              | propyl         |
| A.171 | cyclohexyl                               | propyl         |
| A.172 | trifluoromethyl                          | propyl         |
| A.173 | pentafluoroethyl                         | propyl         |
| A.174 | 4-trifluoromethylphenyl                  | propyl         |
| A.175 | 4-trifluoromethylbenzyl                  | propyl         |
| A.176 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | propyl         |
| A.177 | 2-chloro-1,1-dimethyl-ethyl              | propyl         |
| A.178 | 2-chlorophenyl                           | propyl         |
| A.179 | 2-fluorophenyl                           | propyl         |
| A.180 | 2,4-difluorophenyl                       | propyl         |
| A.181 | 4-chlorophenyl                           | propyl         |
| A.182 | 4-fluorophenyl                           | propyl         |
| A.183 | 2,4-dichlorophenyl                       | propyl         |
| A.184 | 4-trifluoromethoxyphenyl                 | propyl         |
| A.185 | H  | allyl          |
| A.186 | methoxy                                  | allyl          |
| A.187 | ethoxy                                   | allyl          |
| A.188 | n-propoxy                                | allyl          |
| A.189 | n-butoxy                                 | allyl          |
| A.190 | n-pentyloxy                              | allyl          |
| A.191 | n-hexyloxy                               | allyl          |
| A.192 | isopropoxy                               | allyl          |
| A.193 | isobutoxy                                | allyl          |
| A.194 | isopentyloxy                             | allyl          |
| A.195 | tert-butoxy                              | allyl          |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.196 | 2,2-dimethylpropoxy                      | allyl          |
| A.197 | vinyloxy                                 | allyl          |
| A.198 | allyloxy                                 | allyl          |
| A.199 | propargyloxy                             | allyl          |
| A.200 | propen-2-yloxy                           | allyl          |
| A.201 | benzyloxy                                | allyl          |
| A.202 | 4-nitrobenzyloxy                         | allyl          |
| A.203 | methyl                                   | allyl          |
| A.204 | ethyl                                    | allyl          |
| A.205 | n-propyl                                 | allyl          |
| A.206 | n-butyl                                  | allyl          |
| A.207 | n-pentyl                                 | allyl          |
| A.208 | n-hexyl                                  | allyl          |
| A.209 | isopropyl                                | allyl          |
| A.210 | isobutyl                                 | allyl          |
| A.211 | isopentyl                                | allyl          |
| A.212 | tert-butyl                               | allyl          |
| A.213 | 2,2-dimethylpropyl                       | allyl          |
| A.214 | cyclopropyl                              | allyl          |
| A.215 | cyclobutyl                               | allyl          |
| A.216 | cyclopentyl                              | allyl          |
| A.217 | cyclohexyl                               | allyl          |
| A.218 | trifluoromethyl                          | allyl          |
| A.219 | pentafluoroethyl                         | allyl          |
| A.220 | 4-trifluoromethylphenyl                  | allyl          |
| A.221 | 4-trifluoromethylbenzyl                  | allyl          |
| A.222 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | allyl          |
| A.223 | 2-chloro-1,1-dimethyl-ethyl              | allyl          |
| A.224 | 2-chlorophenyl                           | allyl          |
| A.225 | 2-fluorophenyl                           | allyl          |
| A.226 | 2,4-difluorophenyl                       | allyl          |
| A.227 | 4-chlorophenyl                           | allyl          |
| A.228 | 4-fluorophenyl                           | allyl          |
| A.229 | 2,4-dichlorophenyl                       | allyl          |
| A.230 | 4-trifluoromethoxyphenyl                 | allyl          |
| A.231 | H  | propargyl      |
| A.232 | methoxy                                  | propargyl      |
| A.233 | ethoxy                                   | propargyl      |
| A.234 | n-propoxy                                | propargyl      |
| A.235 | n-butoxy                                 | propargyl      |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.236 | n-pentyloxy                              | propargyl      |
| A.237 | n-hexyloxy                               | propargyl      |
| A.238 | isopropoxy                               | propargyl      |
| A.239 | isobutoxy                                | propargyl      |
| A.240 | isopentyloxy                             | propargyl      |
| A.241 | tert-butoxy                              | propargyl      |
| A.242 | 2,2-dimethylpropoxy                      | propargyl      |
| A.243 | vinyloxy                                 | propargyl      |
| A.244 | allyloxy                                 | propargyl      |
| A.245 | propargyloxy                             | propargyl      |
| A.246 | propen-2-yloxy                           | propargyl      |
| A.247 | benzyloxy                                | propargyl      |
| A.248 | 4-nitrobenzyloxy                         | propargyl      |
| A.249 | methyl                                   | propargyl      |
| A.250 | ethyl                                    | propargyl      |
| A.251 | n-propyl                                 | propargyl      |
| A.252 | n-butyl                                  | propargyl      |
| A.253 | n-pentyl                                 | propargyl      |
| A.254 | n-hexyl                                  | propargyl      |
| A.255 | isopropyl                                | propargyl      |
| A.256 | isobutyl                                 | propargyl      |
| A.257 | isopentyl                                | propargyl      |
| A.258 | tert-butyl                               | propargyl      |
| A.259 | 2,2-dimethylpropyl                       | propargyl      |
| A.260 | cyclopropyl                              | propargyl      |
| A.261 | cyclobutyl                               | propargyl      |
| A.262 | cyclopentyl                              | propargyl      |
| A.263 | cyclohexyl                               | propargyl      |
| A.264 | trifluoromethyl                          | propargyl      |
| A.265 | pentafluoroethyl                         | propargyl      |
| A.266 | 4-trifluoromethylphenyl                  | propargyl      |
| A.267 | 4-trifluoromethylbenzyl                  | propargyl      |
| A.268 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | propargyl      |
| A.269 | 2-chloro-1,1-dimethyl-ethyl              | propargyl      |
| A.270 | 2-chlorophenyl                           | propargyl      |
| A.271 | 2-fluorophenyl                           | propargyl      |
| A.272 | 2,4-difluorophenyl                       | propargyl      |
| A.273 | 4-chlorophenyl                           | propargyl      |
| A.274 | 4-fluorophenyl                           | propargyl      |
| A.275 | 2,4-dichlorophenyl                       | propargyl      |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.276 | 4-trifluoromethoxyphenyl                 | propargyl      |
| A.277 | H  | methoxy        |
| A.278 | methoxy                                  | methoxy        |
| A.279 | ethoxy                                   | methoxy        |
| A.280 | n-propoxy                                | methoxy        |
| A.281 | n-butoxy                                 | methoxy        |
| A.282 | n-pentyloxy                              | methoxy        |
| A.283 | n-hexyloxy                               | methoxy        |
| A.284 | isopropoxy                               | methoxy        |
| A.285 | isobutoxy                                | methoxy        |
| A.286 | isopentyloxy                             | methoxy        |
| A.287 | tert-butoxy                              | methoxy        |
| A.288 | 2,2-dimethylpropoxy                      | methoxy        |
| A.289 | vinyloxy                                 | methoxy        |
| A.290 | allyloxy                                 | methoxy        |
| A.291 | propargyloxy                             | methoxy        |
| A.292 | propen-2-yloxy                           | methoxy        |
| A.293 | benzyloxy                                | methoxy        |
| A.294 | 4-nitrobenzyloxy                         | methoxy        |
| A.295 | methyl                                   | methoxy        |
| A.296 | ethyl                                    | methoxy        |
| A.297 | n-propyl                                 | methoxy        |
| A.298 | n-butyl                                  | methoxy        |
| A.299 | n-pentyl                                 | methoxy        |
| A.300 | n-hexyl                                  | methoxy        |
| A.301 | isopropyl                                | methoxy        |
| A.302 | isobutyl                                 | methoxy        |
| A.303 | isopentyl                                | methoxy        |
| A.304 | tert-butyl                               | methoxy        |
| A.305 | 2,2-dimethylpropyl                       | methoxy        |
| A.306 | cyclopropyl                              | methoxy        |
| A.307 | cyclobutyl                               | methoxy        |
| A.308 | cyclopentyl                              | methoxy        |
| A.309 | cyclohexyl                               | methoxy        |
| A.310 | trifluoromethyl                          | methoxy        |
| A.311 | pentafluoroethyl                         | methoxy        |
| A.312 | 4-trifluoromethylphenyl                  | methoxy        |
| A.313 | 4-trifluoromethylbenzyl                  | methoxy        |
| A.314 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | methoxy        |
| A.315 | 2-chloro-1,1-dimethyl-ethyl              | methoxy        |

| No.   | R <sub>9</sub>           | R <sub>5</sub> |
|-------|--------------------------|----------------|
| A.316 | 2-chlorophenyl           | methoxy        |
| A.317 | 2-fluorophenyl           | methoxy        |
| A.318 | 2,4-difluorophenyl       | methoxy        |
| A.319 | 4-chlorophenyl           | methoxy        |
| A.320 | 4-fluorophenyl           | methoxy        |
| A.321 | 2,4-dichlorophenyl       | methoxy        |
| A.322 | 4-trifluoromethoxyphenyl | methoxy        |
| A.323 | H                        | ethoxy         |
| A.324 | methoxy                  | ethoxy         |
| A.325 | ethoxy                   | ethoxy         |
| A.326 | n-propoxy                | ethoxy         |
| A.327 | n-butoxy                 | ethoxy         |
| A.328 | n-pentyloxy              | ethoxy         |
| A.329 | n-hexyloxy               | ethoxy         |
| A.330 | isopropoxy               | ethoxy         |
| A.331 | isobutoxy                | ethoxy         |
| A.332 | isopentyloxy             | ethoxy         |
| A.333 | tert-butoxy              | ethoxy         |
| A.334 | 2,2-dimethylpropoxy      | ethoxy         |
| A.335 | vinyloxy                 | ethoxy         |
| A.336 | allyloxy                 | ethoxy         |
| A.337 | propargyloxy             | ethoxy         |
| A.338 | propen-2-yloxy           | ethoxy         |
| A.339 | benzyloxy                | ethoxy         |
| A.340 | 4-nitrobenzyloxy         | ethoxy         |
| A.341 | methyl                   | ethoxy         |
| A.342 | ethyl                    | ethoxy         |
| A.343 | n-propyl                 | ethoxy         |
| A.344 | n-butyl                  | ethoxy         |
| A.345 | n-pentyl                 | ethoxy         |
| A.346 | n-hexyl                  | ethoxy         |
| A.347 | isopropyl                | ethoxy         |
| A.348 | isobutyl                 | ethoxy         |
| A.349 | isopentyl                | ethoxy         |
| A.350 | tert-butyl               | ethoxy         |
| A.351 | 2,2-dimethylpropyl       | ethoxy         |
| A.352 | cyclopropyl              | ethoxy         |
| A.353 | cyclobutyl               | ethoxy         |
| A.354 | cyclopentyl              | ethoxy         |
| A.355 | cyclohexyl               | ethoxy         |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.356 | trifluoromethyl                          | ethoxy         |
| A.357 | pentafluoroethyl                         | ethoxy         |
| A.358 | 4-trifluoromethylphenyl                  | ethoxy         |
| A.359 | 4-trifluoromethylbenzyl                  | ethoxy         |
| A.360 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | ethoxy         |
| A.361 | 2-chloro-1,1-dimethyl-ethyl              | ethoxy         |
| A.362 | 2-chlorophenyl                           | ethoxy         |
| A.363 | 2-fluorophenyl                           | ethoxy         |
| A.364 | 2,4-difluorophenyl                       | ethoxy         |
| A.365 | 4-chlorophenyl                           | ethoxy         |
| A.366 | 4-fluorophenyl                           | ethoxy         |
| A.367 | 2,4-dichlorophenyl                       | ethoxy         |
| A.368 | 4-trifluoromethoxyphenyl                 | ethoxy         |
| A.369 | H  | methylcarbonyl |
| A.370 | methoxy                                  | methylcarbonyl |
| A.371 | ethoxy                                   | methylcarbonyl |
| A.372 | n-propoxy                                | methylcarbonyl |
| A.373 | n-butoxy                                 | methylcarbonyl |
| A.374 | n-pentyloxy                              | methylcarbonyl |
| A.375 | n-hexyloxy                               | methylcarbonyl |
| A.376 | isopropoxy                               | methylcarbonyl |
| A.377 | isobutoxy                                | methylcarbonyl |
| A.378 | isopentyloxy                             | methylcarbonyl |
| A.379 | tert-butoxy                              | methylcarbonyl |
| A.380 | 2,2-dimethylpropoxy                      | methylcarbonyl |
| A.381 | vinyloxy                                 | methylcarbonyl |
| A.382 | allyloxy                                 | methylcarbonyl |
| A.383 | propargyloxy                             | methylcarbonyl |
| A.384 | propen-2-yloxy                           | methylcarbonyl |
| A.385 | benzyloxy                                | methylcarbonyl |
| A.386 | 4-nitrobenzyloxy                         | methylcarbonyl |
| A.387 | methyl                                   | methylcarbonyl |
| A.388 | ethyl                                    | methylcarbonyl |
| A.389 | n-propyl                                 | methylcarbonyl |
| A.390 | n-butyl                                  | methylcarbonyl |
| A.391 | n-pentyl                                 | methylcarbonyl |
| A.392 | n-hexyl                                  | methylcarbonyl |
| A.393 | isopropyl                                | methylcarbonyl |
| A.394 | isobutyl                                 | methylcarbonyl |
| A.395 | isopentyl                                | methylcarbonyl |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.396 | tert-butyl                               | methylcarbonyl |
| A.397 | 2,2-dimethylpropyl                       | methylcarbonyl |
| A.398 | cyclopropyl                              | methylcarbonyl |
| A.399 | cyclobutyl                               | methylcarbonyl |
| A.400 | cyclopentyl                              | methylcarbonyl |
| A.401 | cyclohexyl                               | methylcarbonyl |
| A.402 | trifluoromethyl                          | methylcarbonyl |
| A.403 | pentafluoroethyl                         | methylcarbonyl |
| A.404 | 4-trifluoromethylphenyl                  | methylcarbonyl |
| A.405 | 4-trifluoromethylbenzyl                  | methylcarbonyl |
| A.406 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | methylcarbonyl |
| A.407 | 2-chloro-1,1-dimethyl-ethyl              | methylcarbonyl |
| A.408 | 2-chlorophenyl                           | methylcarbonyl |
| A.409 | 2-fluorophenyl                           | methylcarbonyl |
| A.410 | 2,4-difluorophenyl                       | methylcarbonyl |
| A.411 | 4-chlorophenyl                           | methylcarbonyl |
| A.412 | 4-fluorophenyl                           | methylcarbonyl |
| A.413 | 2,4-dichlorophenyl                       | methylcarbonyl |
| A.414 | 4-trifluoromethoxyphenyl                 | methylcarbonyl |
| A.415 | H  | ethylcarbonyl  |
| A.416 | methoxy                                  | ethylcarbonyl  |
| A.417 | ethoxy                                   | ethylcarbonyl  |
| A.418 | n-propoxy                                | ethylcarbonyl  |
| A.419 | n-butoxy                                 | ethylcarbonyl  |
| A.420 | n-pentyloxy                              | ethylcarbonyl  |
| A.421 | n-hexyloxy                               | ethylcarbonyl  |
| A.422 | isopropoxy                               | ethylcarbonyl  |
| A.423 | isobutoxy                                | ethylcarbonyl  |
| A.424 | isopentyloxy                             | ethylcarbonyl  |
| A.425 | tert-butoxy                              | ethylcarbonyl  |
| A.426 | 2,2-dimethylpropoxy                      | ethylcarbonyl  |
| A.427 | vinyloxy                                 | ethylcarbonyl  |
| A.428 | allyloxy                                 | ethylcarbonyl  |
| A.429 | propargyloxy                             | ethylcarbonyl  |
| A.430 | propen-2-yloxy                           | ethylcarbonyl  |
| A.431 | benzyloxy                                | ethylcarbonyl  |
| A.432 | 4-nitrobenzyloxy                         | ethylcarbonyl  |
| A.433 | methyl                                   | ethylcarbonyl  |
| A.434 | ethyl                                    | ethylcarbonyl  |
| A.435 | n-propyl                                 | ethylcarbonyl  |

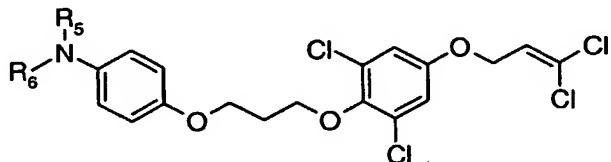
| No.   | R <sub>9</sub>                           | R <sub>5</sub>      |
|-------|--|---------------------|
| A.436 | n-butyl                                  | ethylcarbonyl       |
| A.437 | n-pentyl                                 | ethylcarbonyl       |
| A.438 | n-hexyl                                  | ethylcarbonyl       |
| A.439 | isopropyl                                | ethylcarbonyl       |
| A.440 | isobutyl                                 | ethylcarbonyl       |
| A.441 | isopentyl                                | ethylcarbonyl       |
| A.442 | tert-butyl                               | ethylcarbonyl       |
| A.443 | 2,2-dimethylpropyl                       | ethylcarbonyl       |
| A.444 | cyclopropyl                              | ethylcarbonyl       |
| A.445 | cyclobutyl                               | ethylcarbonyl       |
| A.446 | cyclopentyl                              | ethylcarbonyl       |
| A.447 | cyclohexyl                               | ethylcarbonyl       |
| A.448 | trifluoromethyl                          | ethylcarbonyl       |
| A.449 | pentafluoroethyl                         | ethylcarbonyl       |
| A.450 | 4-trifluoromethylphenyl                  | ethylcarbonyl       |
| A.451 | 4-trifluoromethylbenzyl                  | ethylcarbonyl       |
| A.452 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | ethylcarbonyl       |
| A.453 | 2-chloro-1,1-dimethyl-ethyl              | ethylcarbonyl       |
| A.454 | 2-chlorophenyl                           | ethylcarbonyl       |
| A.455 | 2-fluorophenyl                           | ethylcarbonyl       |
| A.456 | 2,4-difluorophenyl                       | ethylcarbonyl       |
| A.457 | 4-chlorophenyl                           | ethylcarbonyl       |
| A.458 | 4-fluorophenyl                           | ethylcarbonyl       |
| A.459 | 2,4-dichlorophenyl                       | ethylcarbonyl       |
| A.460 | 4-trifluoromethoxyphenyl                 | ethylcarbonyl       |
| A.461 | H  | cyclopropylcarbonyl |
| A.462 | methoxy                                  | cyclopropylcarbonyl |
| A.463 | ethoxy                                   | cyclopropylcarbonyl |
| A.464 | n-propoxy                                | cyclopropylcarbonyl |
| A.465 | n-butoxy                                 | cyclopropylcarbonyl |
| A.466 | n-pentyloxy                              | cyclopropylcarbonyl |
| A.467 | n-hexyloxy                               | cyclopropylcarbonyl |
| A.468 | isopropoxy                               | cyclopropylcarbonyl |
| A.469 | isobutoxy                                | cyclopropylcarbonyl |
| A.470 | isopentyloxy                             | cyclopropylcarbonyl |
| A.471 | tert-butoxy                              | cyclopropylcarbonyl |
| A.472 | 2,2-dimethylpropoxy                      | cyclopropylcarbonyl |
| A.473 | vinyloxy                                 | cyclopropylcarbonyl |
| A.474 | allyloxy                                 | cyclopropylcarbonyl |
| A.475 | propargyloxy                             | cyclopropylcarbonyl |

| No.   | R <sub>9</sub>                           | R <sub>5</sub>      |
|-------|--|---------------------|
| A.476 | propen-2-yloxy                           | cyclopropylcarbonyl |
| A.477 | benzyloxy                                | cyclopropylcarbonyl |
| A.478 | 4-nitrobenzyloxy                         | cyclopropylcarbonyl |
| A.479 | methyl                                   | cyclopropylcarbonyl |
| A.480 | ethyl                                    | cyclopropylcarbonyl |
| A.481 | n-propyl                                 | cyclopropylcarbonyl |
| A.482 | n-butyl                                  | cyclopropylcarbonyl |
| A.483 | n-pentyl                                 | cyclopropylcarbonyl |
| A.484 | n-hexyl                                  | cyclopropylcarbonyl |
| A.485 | isopropyl                                | cyclopropylcarbonyl |
| A.486 | isobutyl                                 | cyclopropylcarbonyl |
| A.487 | isopentyl                                | cyclopropylcarbonyl |
| A.488 | tert-butyl                               | cyclopropylcarbonyl |
| A.489 | 2,2-dimethylpropyl                       | cyclopropylcarbonyl |
| A.490 | cyclopropyl                              | cyclopropylcarbonyl |
| A.491 | cyclobutyl                               | cyclopropylcarbonyl |
| A.492 | cyclopentyl                              | cyclopropylcarbonyl |
| A.493 | cyclohexyl                               | cyclopropylcarbonyl |
| A.494 | trifluoromethyl                          | cyclopropylcarbonyl |
| A.495 | pentafluoroethyl                         | cyclopropylcarbonyl |
| A.496 | 4-trifluoromethylphenyl                  | cyclopropylcarbonyl |
| A.497 | 4-trifluoromethylbenzyl                  | cyclopropylcarbonyl |
| A.498 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | cyclopropylcarbonyl |
| A.499 | 2-chloro-1,1-dimethyl-ethyl              | cyclopropylcarbonyl |
| A.500 | 2-chlorophenyl                           | cyclopropylcarbonyl |
| A.501 | 2-fluorophenyl                           | cyclopropylcarbonyl |
| A.502 | 2,4-difluorophenyl                       | cyclopropylcarbonyl |
| A.503 | 4-chlorophenyl                           | cyclopropylcarbonyl |
| A.504 | 4-fluorophenyl                           | cyclopropylcarbonyl |
| A.505 | 2,4-dichlorophenyl                       | cyclopropylcarbonyl |
| A.506 | 4-trifluoromethoxyphenyl                 | cyclopropylcarbonyl |
| A.507 | H  | methoxycarbonyl     |
| A.508 | methoxy                                  | methoxycarbonyl     |
| A.509 | ethoxy                                   | methoxycarbonyl     |
| A.510 | n-propoxy                                | methoxycarbonyl     |
| A.511 | n-butoxy                                 | methoxycarbonyl     |
| A.512 | n-pentyloxy                              | methoxycarbonyl     |
| A.513 | n-hexyloxy                               | methoxycarbonyl     |
| A.514 | isopropoxy                               | methoxycarbonyl     |
| A.515 | isobutoxy                                | methoxycarbonyl     |

| No.   | R <sub>9</sub>                           | R <sub>5</sub>  |
|-------|--|-----------------|
| A.516 | isopentyloxy                             | methoxycarbonyl |
| A.517 | tert-butoxy                              | methoxycarbonyl |
| A.518 | 2,2-dimethylpropoxy                      | methoxycarbonyl |
| A.519 | vinyloxy                                 | methoxycarbonyl |
| A.520 | allyloxy                                 | methoxycarbonyl |
| A.521 | propargyloxy                             | methoxycarbonyl |
| A.522 | propen-2-yloxy                           | methoxycarbonyl |
| A.523 | benzyloxy                                | methoxycarbonyl |
| A.524 | 4-nitrobenzyloxy                         | methoxycarbonyl |
| A.525 | methyl                                   | methoxycarbonyl |
| A.526 | ethyl                                    | methoxycarbonyl |
| A.527 | n-propyl                                 | methoxycarbonyl |
| A.528 | n-butyl                                  | methoxycarbonyl |
| A.529 | n-pentyl                                 | methoxycarbonyl |
| A.530 | n-hexyl                                  | methoxycarbonyl |
| A.531 | isopropyl                                | methoxycarbonyl |
| A.532 | isobutyl                                 | methoxycarbonyl |
| A.533 | isopentyl                                | methoxycarbonyl |
| A.534 | tert-butyl                               | methoxycarbonyl |
| A.535 | 2,2-dimethylpropyl                       | methoxycarbonyl |
| A.536 | cyclopropyl                              | methoxycarbonyl |
| A.537 | cyclobutyl                               | methoxycarbonyl |
| A.538 | cyclopentyl                              | methoxycarbonyl |
| A.539 | cyclohexyl                               | methoxycarbonyl |
| A.540 | trifluoromethyl                          | methoxycarbonyl |
| A.541 | pentafluoroethyl                         | methoxycarbonyl |
| A.542 | 4-trifluoromethylphenyl                  | methoxycarbonyl |
| A.543 | 4-trifluoromethylbenzyl                  | methoxycarbonyl |
| A.544 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | methoxycarbonyl |
| A.545 | 2-chloro-1,1-dimethyl-ethyl              | methoxycarbonyl |
| A.546 | 2-chlorophenyl                           | methoxycarbonyl |
| A.547 | 2-fluorophenyl                           | methoxycarbonyl |
| A.548 | 2,4-difluorophenyl                       | methoxycarbonyl |
| A.549 | 4-chlorophenyl                           | methoxycarbonyl |
| A.550 | 4-fluorophenyl                           | methoxycarbonyl |
| A.551 | 2,4-dichlorophenyl                       | methoxycarbonyl |
| A.552 | 4-trifluoromethoxyphenyl                 | methoxycarbonyl |
| A.553 | H  | ethoxycarbonyl  |
| A.554 | methoxy                                  | ethoxycarbonyl  |
| A.555 | ethoxy                                   | ethoxycarbonyl  |

| No.   | R <sub>9</sub>                           | R <sub>5</sub> |
|-------|--|----------------|
| A.556 | n-propoxy                                | ethoxycarbonyl |
| A.557 | n-butoxy                                 | ethoxycarbonyl |
| A.558 | n-pentyloxy                              | ethoxycarbonyl |
| A.559 | n-hexyloxy                               | ethoxycarbonyl |
| A.560 | isopropoxy                               | ethoxycarbonyl |
| A.561 | isobutoxy                                | ethoxycarbonyl |
| A.562 | isopentyloxy                             | ethoxycarbonyl |
| A.563 | tert-butoxy                              | ethoxycarbonyl |
| A.564 | 2,2-dimethylpropoxy                      | ethoxycarbonyl |
| A.565 | Vinyloxy                                 | ethoxycarbonyl |
| A.566 | Allyloxy                                 | ethoxycarbonyl |
| A.567 | propargyloxy                             | ethoxycarbonyl |
| A.568 | propen-2-yloxy                           | ethoxycarbonyl |
| A.569 | Benzylxy                                 | ethoxycarbonyl |
| A.570 | 4-nitrobenzylxy                          | ethoxycarbonyl |
| A.571 | Methyl                                   | ethoxycarbonyl |
| A.572 | Ethyl                                    | ethoxycarbonyl |
| A.573 | n-propyl                                 | ethoxycarbonyl |
| A.574 | n-butyl                                  | ethoxycarbonyl |
| A.575 | n-pentyl                                 | ethoxycarbonyl |
| A.576 | n-hexyl                                  | ethoxycarbonyl |
| A.577 | Isopropyl                                | ethoxycarbonyl |
| A.578 | Isobutyl                                 | ethoxycarbonyl |
| A.579 | isopentyl                                | ethoxycarbonyl |
| A.580 | tert-butyl                               | ethoxycarbonyl |
| A.581 | 2,2-dimethylpropyl                       | ethoxycarbonyl |
| A.582 | cyclopropyl                              | ethoxycarbonyl |
| A.583 | cyclobutyl                               | ethoxycarbonyl |
| A.584 | cyclopentyl                              | ethoxycarbonyl |
| A.585 | cyclohexyl                               | ethoxycarbonyl |
| A.586 | trifluoromethyl                          | ethoxycarbonyl |
| A.587 | pentafluoroethyl                         | ethoxycarbonyl |
| A.588 | 4-trifluoromethylphenyl                  | ethoxycarbonyl |
| A.589 | 4-trifluoromethylbenzyl                  | ethoxycarbonyl |
| A.590 | 2-chloro-(1-chloromethyl-1-methyl)-ethyl | ethoxycarbonyl |
| A.591 | 2-chloro-1,1-dimethyl-ethyl              | ethoxycarbonyl |
| A.592 | 2-chlorophenyl                           | ethoxycarbonyl |
| A.593 | 2-fluorophenyl                           | ethoxycarbonyl |
| A.594 | 2,4-difluorophenyl                       | ethoxycarbonyl |
| A.595 | 4-chlorophenyl                           | ethoxycarbonyl |

| No.   | R <sub>9</sub>           | R <sub>5</sub> |
|-------|--------------------------|----------------|
| A.596 | 4-fluorophenyl           | ethoxycarbonyl |
| A.597 | 2,4-dichlorophenyl       | ethoxycarbonyl |
| A.598 | 4-trifluoromethoxyphenyl | ethoxycarbonyl |

**Table 1:** Compounds of formula

| No. | R <sub>5</sub>                 | R <sub>6</sub>  | <sup>1</sup> H-NMR (CDCl <sub>3</sub> ) 300MHz; m.p. (°C)  |
|-----|--------------------------------|-----------------|--|
| 1.1 | H                              | H               | 2.26 (m,2H), 3.40 (s,NH <sub>2</sub> ), 4.10-4.22 (m,6H), 4.59 (d,2H), 6.12 (t,1H), 6.63 (d,2H), 6.78 (d,2H), 6.82 (s,2H)                            |
| 1.2 | methyl                         | CH <sub>3</sub> | 2.26 (m,2H), 2.88 (s,6H) 4.10-4.22 (m,4H), 4.58 (d,2H), 6.10 (t,1H), 6.74 (d,2H), 6.82 (s,2H), 6.88 (d,2H)   |
| 1.3 | methoxycarbonyl                | H               | 2.28 (m,2H), 3.76 (s,3H), 4.13 (t,2H), 4.22 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.50 (s,NH), 6.82 (s,2H), 6.88 (d,2H), 7.28 (d,2H)                     |
| 1.4 | ethoxycarbonyl                 | H               | 1.31 (t,3H), 2.28 (m,2H), 4.17 (t,2H), 4.20-4.30 (t,2H+q,2H), 4.58 (d,2H), 6.12 (t,1H), 6.47 (s,NH), 6.85 (s,2H), 6.88 (d,2H), 7.28 (d,2H)           |
| 1.5 | isopropoxycarbonyl             | H               | 1.30 (d,6H), 2.28 (m,2H), 4.15 (t,2H), 4.23 (t,2H), 4.58 (d,2H), 5.02 (m,1H), 6.11 (t,1H), 6.40 (s,NH), 6.85 (s,2H), 6.89 (d,2H), 7.28 (d,2H)        |
| 1.6 | tert-butyloxycarbonyl          | H               | 1.50 (s,9H), 2.28 (m,2H), 4.13 (t,2H), 4.21 (t,2H), 4.58 (d,2H), 6.12 (t,1H), 6.37 (s,NH), 6.82 (s,2H), 6.88 (d,2H), 7.28 (d,2H)                     |
| 1.7 | 2,2-dimethyl-propyloxycarbonyl | H               | 0.98 (s,9H), 2.29 (m,2H), 3.38 (s,2H), 4.16 (t,2H), 4.23 (t,2H), 4.58 (d,2H), 6.11 (t,1H), 6.50 (s,NH), 6.82 (s,2H), 6.89 (d,2H), 7.28 (d,2H)        |
| 1.8 | vinyloxycarbonyl               | H               | 2.29 (m,2H), 4.17 (t,2H), 4.24 (t,2H), 4.52 (d,1H), 4.58 (d,2H) 4.83 (d,1H), 6.12 (t,1H), 6.63 (s,NH), 6.86 (s,2H), 6.92 (d,2H), 7.25-7.37 (m,2H+1H) |

| No.  | R <sub>5</sub>                      | R <sub>6</sub> | <sup>1</sup> H-NMR (CDCl <sub>3</sub> ) 300MHz; m.p. (°C)   |
|------|-------------------------------------|----------------|---|
| 1.9  | allyloxycarbonyl                    | H              | 2.29 (m,2H), 4.17 (t,2H), 4.23 (t,2H), 4.59 (d,2H) 4.66 (d,2H), 5.27 (d,1H), 5.37 (d,1H) 5.9-6.09 (m,1H), 6.12 (t,1H), 6.51 (s,NH), 6.85 (s,2H), 6.90 (d,2H), 7.29 (d,2H) |
| 1.10 | propargyloxycarbonyl                | H              | 2.28 (m,2H), 2.51 (s,1H), 4.15 (t,2H), 4.25 (t,2H), 4.60 (d,2H), 4.80 (s,2H), 6.11 (t,1H), 6.63 (s,NH), 6.88 (s,2H), 6.90 (d,2H), 7.30 (d,2H)                             |
| 1.11 | propen-2-yloxycarbonyl              | H              | 2.00 (s,3H), 2.29 (m,2H), 4.17 (t,2H), 4.23 (t,2H), 4.52 (dd,1H), 4.49 (d,2H) 4.72 (s,1H), 4.80 (s,1H) 6.12 (t,1H), 6.60 (s,NH), 6.83 (s,2H), 6.91 (d,2H), 7.32 (d,2H)    |
| 1.12 | benzyloxycarbonyl                   | H              | 2.28 (m,2H), 4.17 (t,2H), 4.25 (t,2H), 4.60 (d,2H), 5.19 (s,2H), 6.12 (t,1H), 6.54 (s,NH), 6.83 (s,2H), 6.90 (d,2H), 7.28-7.45 (m,7H)                                     |
| 1.13 | 4-nitrobenzyloxycarbonyl            | H              | 2.28 (m,2H), 4.13 (t,2H), 4.23 (t,2H), 4.59 (d,2H), 5.30 (s,2H), 6.12 (t,1H), 6.62 (s,NH), 6.83 (s,2H), 6.90 (d,2H), 7.39 (d,2H), 7.57 (d,2H), 8.23 (d,2H)                |
| 1.14 | methylcarbonyl                      | H              | 2.18 (s,3H), 2.29 (m,2H), 4.13 (t,2H), 4.23 (t,2H), 4.58 (d,2H), 6.12 (t,1H), 6.83 (s,2H), 6.90 (d,2H), 7.03 (s,NH), 7.40 (d,2H)  |
| 1.15 | 4-trifluoromethylcarbonyl           | H              | 2.39 (m,2H), 4.17 (t,2H), 4.28 (t,2H), 4.58 (d,2H), 6.12 (t,1H), 6.83 (s,2H), 6.94 (d,2H), 7.47 (d,2H), 7.79 (s,NH)   |
| 1.16 | 4-trifluoromethylphenyl-carbonyl    | H              | 2.29 (m,2H), 4.16 (t,2H), 4.28 (t,2H), 4.58 (d,2H), 6.11 (t,1H), 6.83 (s,2H), 6.93 (d,2H), 7.53 (d,2H), 7.72 (d,2H), 7.88 (s,NH), 7.98 (d,2H)                             |
| 1.17 | 4-trifluoromethylbenzyl-carbonyl    | H              | 2.25 (m,2H), 3.77 (s,2H), 4.13 (t,2H), 4.21 (t,2H), 4.58 (d,2H), 6.11 (t,1H), 6.82 (s,2H), 6.87 (d,2H), 7.18 (s,NH), 7.33 (d,2H), 7.48 (d,2H), 7.63 (d,2H)                |
| 1.18 | 2-chloro-1,1-dimethyl-ethylcarbonyl | H              | 1.40 (s,6H), 2.29 (m,2H), 3.70 (s,2H), 4.13 (t,2H), 4.23 (t,2H), 4.57 (d,2H), 6.10 (t,1H), 6.82 (s,2H), 6.90 (d,2H), 7.37-7.45 (d,2H+s,NH)                                |
| 1.19 | 2-chlorobenzoyl                     | H              | m.p.: 107-111°C   |
| 1.20 | 2-toluoyl                           | H              | m.p.: 89-92°C   |
| 1.21 | 2-bromobenzoyl                      | H              | m.p.: 112-114°C   |
| 1.22 | 2,6-difluorobenzoyl                 | H              | m.p.: 95-97°C   |
| 1.23 | 2,4-difluorobenzoyl                 | H              | m.p.: 89-91°C   |
| 1.24 | 2-trifluoromethylbenzoyl            | H              | m.p.: 118-121°C   |

| No.  | R <sub>5</sub>         | R <sub>6</sub> | <sup>1</sup> H-NMR (CDCl <sub>3</sub> ) 300MHz; m.p. (°C) |
|------|------------------------|----------------|---|
| 1.25 | 2-furoyl               | H              | m.p.: 108-113°C   |
| 1.26 | 2-thenoyl              | H              | m.p.: 83-90°C   |
| 1.27 | Ethoxycarbonyl         | Ethyl          | n <sub>D</sub> <sup>20</sup> : 1.5581                     |
| 1.28 | tert.-butyloxycarbonyl | Ethyl          | Resin   |

Table 2: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 3: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 4: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 5: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 6: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 7: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 8: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 9: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 10: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, A<sub>1</sub> is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 11: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 12: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 13: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 14: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 15: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 16: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 17: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 18: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 19: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 20: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 21: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

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Table 22: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 23: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 24: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 25: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 26: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 27: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 28: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is a bond, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 29: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 30: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 31: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 32: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 33: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 34: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 35: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 36: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 37: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 38: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 39: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 40: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 41: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 42: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 43: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 44: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 45: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 46: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 47: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 48: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 49: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 50: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 51: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 52: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 53: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 54: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 55: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 56: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 57: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 58: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 2, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 59: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

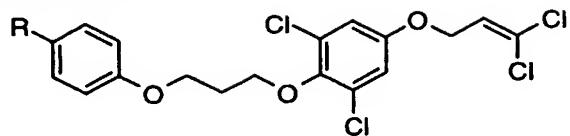
Table 60: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 61: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 3, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

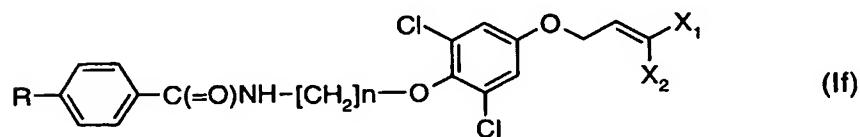
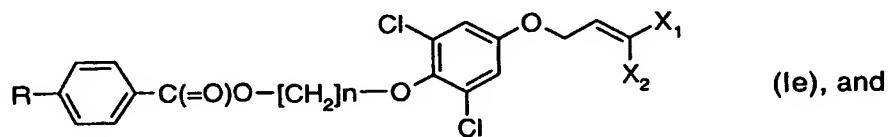
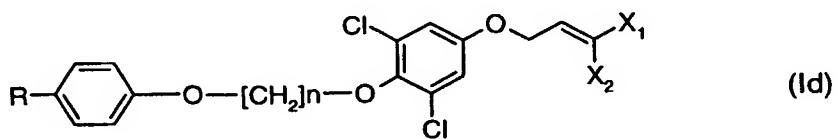
Table 62: Compounds of general formula (Ia) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 63: Compounds of general formula (Ib) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

Table 64: Compounds of general formula (Ic) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, A1 is -CH<sub>2</sub>-CH<sub>2</sub>-, n is 4, and the combination of substituents R<sub>5</sub> and R<sub>9</sub> for each compound corresponds to a line A.1 to A.598 of Table A.

**Table 65:** Compounds of formula

| No.  | R | $^1\text{H-NMR}$ ( $\text{CDCl}_3$ ) 300MHz   |
|------|---|---|
| 93.1 |   | 2.13 (m,2H), 2.28 (m,2H), 2.59 (t,2H), 3.82 (t,2H), 4.14 (t,2H), 4.25 (t,2H), 4.58 (d,2H), 6.11 (t,1H), 6.82 (s,2H), 6.92 (d,2H), 7.50 (d,2H) |
| 93.2 |   | 2.29 (m,2H), 2.89 (s,4H), 4.13 (t,2H), 4.28 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.82 (s,2H), 7.01 (d,2H), 7.18 (d,2H)                           |
| 93.3 |   | 2.29 (m,2H), 4.02 (t,2H), 4.15 (t,2H), 4.25 (t,2H), 4.47 (t,2H), 4.58 (d,2H), 6.11 (t,1H), 6.82 (s,2H), 6.93 (d,2H), 7.43 (d,2H)              |
| 93.4 |   |   |
| 93.5 |   |   |

**Table B:** Compounds of formulae

| No.  | R  |
|------|--|
| B.1  | 3-(oxazolidin)-yl-2-one                      |
| B.2  | 3-(4-methyl-oxazolidin)-yl-2-one             |
| B.3  | 3-(4-ethyl-oxazolidin)-yl-2-one              |
| B.4  | 3-(4-propyl-oxazolidin)-yl-2-one             |
| B.5  | 3-(4,4-dimethyl-oxazolidin)-yl-2-one         |
| B.6  | 3-(4,4-Diethyl-oxazolidin)-yl-2-one          |
| B.7  | 3-(5-Methyl-oxazolidin)-yl-2-one             |
| B.8  | 3-(5-Ethyl-oxazolidin)-yl-2-one              |
| B.9  | 3-(5-Propyl-oxazolidin)-yl-2-one             |
| B.10 | 3-(5-Cyclopropyl-oxazolidin)-yl-2-one        |
| B.11 | 3-(5-Isopropyl-oxazolidin)-yl-2-one          |
| B.12 | 3-(5-Isobutyl-oxazolidin)-yl-2-one           |
| B.13 | 3-(5-Allyl-oxazolidin)-yl-2-one              |
| B.14 | 3-(5-Methoxymethyl-oxazolidin)-yl-2-one      |
| B.15 | 3-(5-Ethoxymethyl-oxazolidin)-yl-2-one       |
| B.16 | 3-(5-Propoxymethyl-oxazolidin)-yl-2-one      |
| B.17 | 3-(5-Cyclopropoxymethyl-oxazolidin)-yl-2-one |
| B.18 | 3-(5-Isopropoxymethyl-oxazolidin)-yl-2-one   |
| B.19 | 3-(5-isobutoxymethyl-oxazolidin)-yl-2-one    |
| B.20 | 3-(5-allyloxymethyl-oxazolidin)-yl-2-one     |
| B.21 | 3-(5-propargyloxymethyl-oxazolidin)-yl-2-one |
| B.22 | 3-(5,5-dimethyl-oxazolidin)-yl-2-one         |
| B.23 | 3-(5,5-diethyl-oxazolidin)-yl-2-one          |
| B.24 | 1-(pyrrolidin)-yl-2-one                      |
| B.25 | 1-(3-methyl-pyrrolidin)-yl-2-one             |
| B.26 | 1-(3-ethyl-pyrrolidin)-yl-2-one              |
| B.27 | 1-(3-propyl-pyrrolidin)-yl-2-one             |
| B.28 | 1-(3-cyclopropyl-pyrrolidin)-yl-2-one        |
| B.29 | 1-(3-isopropyl-pyrrolidin)-yl-2-one          |
| B.30 | 1-(3-isobutyl-pyrrolidin)-yl-2-one           |
| B.31 | 1-(3-allyl-pyrrolidin)-yl-2-one              |
| B.32 | 1-(3-methoxymethyl-pyrrolidin)-yl-2-one      |
| B.33 | 1-(3-ethoxymethyl-pyrrolidin)-yl-2-one       |
| B.34 | 1-(3-propoxymethyl-pyrrolidin)-yl-2-one      |
| B.35 | 1-(3-cyclopropoxymethyl-pyrrolidin)-yl-2-one |
| B.36 | 1-(3-isopropoxymethyl-pyrrolidin)-yl-2-one   |
| B.37 | 1-(3-isobutoxymethyl-pyrrolidin)-yl-2-one    |
| B.38 | 1-(3-allyloxymethyl-pyrrolidin)-yl-2-one     |

| No.  | R  |
|------|--|
| B.39 | 1-(3,3-dimethyl-pyrrolidin)-yl-2-one             |
| B.40 | 1-(3,3-diethyl-pyrrolidin)-yl-2-one              |
| B.41 | 1-(5-methyl-pyrrolidin)-yl-2-one                 |
| B.42 | 1-(5-ethyl-pyrrolidin)-yl-2-one                  |
| B.43 | 1-(5-propyl-pyrrolidin)-yl-2-one                 |
| B.44 | 1-(5-cyclopropyl-pyrrolidin)-yl-2-one            |
| B.45 | 1-(5-isopropyl-pyrrolidin)-yl-2-one              |
| B.46 | 1-(5-isobutyl-pyrrolidin)-yl-2-one               |
| B.47 | 1-(5-allyl-pyrrolidin)-yl-2-one                  |
| B.48 | 1-(5-methoxymethyl-pyrrolidin)-yl-2-one          |
| B.49 | 1-(5-ethoxymethyl-pyrrolidin)-yl-2-one           |
| B.50 | 1-(5-propoxymethyl-pyrrolidin)-yl-2-one          |
| B.51 | 1-(5-cyclopropoxymethyl-pyrrolidin)-yl-2-one     |
| B.52 | 1-(5-isopropoxymethyl-pyrrolidin)-yl-2-one       |
| B.53 | 1-(5-isobutoxymethyl-pyrrolidin)-yl-2-one        |
| B.54 | 1-(5-allyloxymethyl-pyrrolidin)-yl-2-one         |
| B.55 | 1-(5,5-dimethyl-pyrrolidin)-yl-2-one             |
| B.56 | 1-(5,5-diethyl-pyrrolidin)-yl-2-one              |
| B.57 | 1-(pyrrolidin)-yl-2,5-dione                      |
| B.58 | 1-(3-methyl-pyrrolidin)-yl-2,5-dione             |
| B.59 | 1-(3-ethyl-pyrrolidin)-yl-2,5-dione              |
| B.60 | 1-(3-propyl-pyrrolidin)-yl-2,5-dione             |
| B.61 | 1-(3-cyclopropyl-pyrrolidin)-yl-2,5-dione        |
| B.62 | 1-(3-isopropyl-pyrrolidin)-yl-2,5-dione          |
| B.63 | 1-(3-isobutyl-pyrrolidin)-yl-2,5-dione           |
| B.64 | 1-(3-allyl-pyrrolidin)-yl-2,5-dione              |
| B.65 | 1-(3-methoxymethyl-pyrrolidin)-yl-2,5-dione      |
| B.66 | 1-(3-ethoxymethyl-pyrrolidin)-yl-2,5-dione       |
| B.67 | 1-(3-propoxymethyl-pyrrolidin)-yl-2,5-dione      |
| B.68 | 1-(3-cyclopropoxymethyl-pyrrolidin)-yl-2,5-dione |
| B.69 | 1-(3-isopropoxymethyl-pyrrolidin)-yl-2,5-dione   |
| B.70 | 1-(3-isobutoxymethyl-pyrrolidin)-yl-2,5-dione    |
| B.71 | 1-(3-allyloxymethyl-pyrrolidin)-yl-2,5-dione     |
| B.72 | 1-(3,3-dimethyl-pyrrolidin)-yl-2,5-dione         |
| B.73 | 1-(3,3-diethyl-pyrrolidin)-yl-2,5-dione          |
| B.74 | 1-(3-methyl-pyrrolin)-yl-2,5-dione               |
| B.75 | 1-(3-ethyl-pyrrolin)-yl-2,5-dione                |
| B.76 | 1-(3-n-propyl-pyrrolin)-yl-2,5-dione             |
| B.77 | 1-(3,4-dimethyl-pyrrolin)-yl-2,5-dione           |
| B.78 | 1-(3,4-diethyl-pyrrolin)-yl-2,5-dione            |

| No.  | R   |
|------|---|
| B.79 | 1-(3,4-di-n-propyl-pyrrolin)-yl-2,5-dione |
| B.80 | 1-(3-acetyl-pyrrolin)-yl-2,5-dione        |
| B.81 | 1-(3-n-propionyl-pyrrolin)-yl-2,5-dione   |
| B.82 | 1-(3-cyano-pyrrolin)-yl-2,5-dione         |

Table 66: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 67: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 68: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 69: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 70: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 71: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 72: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 73: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 74: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are fluorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 75: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 76: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 77: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 78: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 79: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 80: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 81: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 82: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 83: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 84: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 85: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 86: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 2, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 87: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 88: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

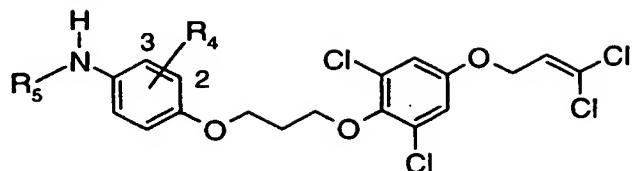
Table 89: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 3, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 90: Compounds of general formula (Id) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

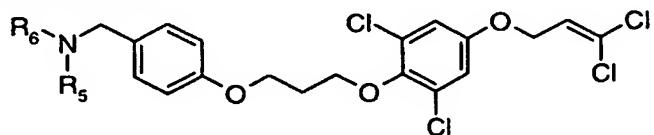
Table 91: Compounds of general formula (Ie) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

Table 92: Compounds of general formula (If) wherein X<sub>1</sub> and X<sub>2</sub> are bromine, n is 4, and the substituent R for each compound corresponds to a line B.1 to B.82 of Table B.

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Table 93: Compounds of formula

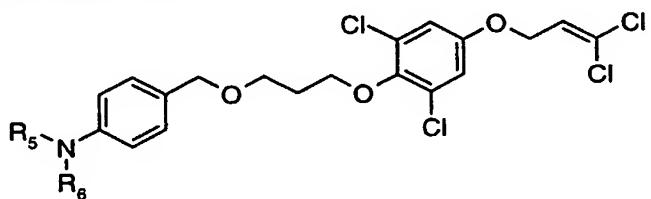
| No.   | R <sub>5</sub>          | R <sub>4</sub>       | Physical data                         |
|-------|-------------------------|----------------------|---------------------------------------|
| 93.6  | Ethoxycarbonyl          | 3-methyl             | Resin                                 |
| 93.7  | Tert-butyloxycarbonyl   | 2,3-dimethyl         | Resin                                 |
| 93.8  | Tert-butyloxycarbonyl   | 3-chloro             | n <sub>D</sub> <sup>20</sup> : 1.5651 |
| 93.9  | Tert-butyloxycarbonyl   | 3-fluoro             | n <sub>D</sub> <sup>20</sup> : 1.5545 |
| 93.10 | Tert-butyloxycarbonyl   | 2,6-dibromo          | Resin                                 |
| 93.11 | Tert-butyloxycarbonyl   | 2,6-dichloro         | Resin                                 |
| 93.12 | Tert-butyloxycarbonyl   | 2-isopropyl-5-methyl | Resin                                 |
| 93.13 | Tert-butyloxycarbonyl   | 2-methoxy            | Resin                                 |
| 93.14 | Tert-butyloxycarbonyl   | 2-methyl             | Resin                                 |
| 93.15 | Tert-butyloxycarbonyl   | 3-methyl             | Resin                                 |
| 93.16 | isopropoxycarbonyl      | 3-methyl             | Resin                                 |
| 93.17 | Tert-butyloxycarbonyl   | 2-nitro              | Resin                                 |
| 93.18 | Tert-butyloxycarbonyl   | 3-nitro              | n <sub>D</sub> <sup>20</sup> : 1.5976 |
| 93.19 | Methylcarbonyl          | 3,5-dichloro         |                                       |
| 93.20 | Ethylcarbonyl           | 3,5-dichloro         |                                       |
| 93.21 | Isopropylcarbonyl       | 3,5-dichloro         |                                       |
| 93.22 | Tert-butylcarbonyl      | 3,5-dichloro         |                                       |
| 93.23 | 2-Chlorophenyl-carbonyl | 3,5-dichloro         |                                       |
| 93.24 | 4-Fluorophenyl-carbonyl | 3,5-dichloro         |                                       |
| 93.25 | 2-Methylphenyl-carbonyl | 3,5-dichloro         |                                       |
| 93.26 | Methoxycarbonyl         | 3,5-dichloro         |                                       |
| 93.27 | Ethoxycarbonyl          | 3,5-dichloro         |                                       |
| 93.28 | Isopropoxycarbonyl      | 3,5-dichloro         |                                       |
| 93.29 | Tert-butyloxycarbonyl   | 3,5-dichloro         |                                       |

Table 94: Compounds of formula

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| No.   | R <sub>5</sub>         | R <sub>6</sub> | physical data                         |
|-------|------------------------|----------------|---------------------------------------|
| 94.1  | Methoxycarbonyl        | Methyl         | n <sub>D</sub> <sup>20</sup> : 1.5646 |
| 94.2  | Ethoxycarbonyl         | Methyl         | Resin                                 |
| 94.3  | Isopropoxycarbonyl     | Methyl         | n <sub>D</sub> <sup>20</sup> : 1.5511 |
| 94.4  | 58.methoxycarbonyl     | ethyl          | n <sub>D</sub> <sup>20</sup> : 1.5620 |
| 94.5  | Ethoxycarbonyl         | ethyl          | n <sub>D</sub> <sup>20</sup> : 1.5548 |
| 94.6  | Isopropoxycarbonyl     | ethyl          | n <sub>D</sub> <sup>20</sup> : 1.5430 |
| 94.7  | Methoxycarbonyl        | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5572 |
| 94.8  | Ethoxycarbonyl         | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5505 |
| 94.9  | Isopropoxycarbonyl     | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5450 |
| 94.10 | Propargyloxycarbonyl   | methyl         | n <sub>D</sub> <sup>20</sup> : 1.5677 |
| 94.11 | Allyloxycarbonyl       | methyl         | n <sub>D</sub> <sup>20</sup> : 1.5639 |
| 94.12 | Trifluormethylcarbonyl | methyl         | m.p.: 84-88°C                         |
| 94.13 | 2-chlorbenzoyl         | methyl         | n <sub>D</sub> <sup>20</sup> : 1.5887 |
| 94.14 | 2-fluorbenzoyl         | methyl         | n <sub>D</sub> <sup>20</sup> : 1.5817 |
| 94.15 | 2,4-difluorbenzoyl     | methyl         | n <sub>D</sub> <sup>20</sup> : 1.5771 |
| 94.16 | Propargyloxycarbonyl   | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5606 |
| 94.17 | Allyloxycarbonyl       | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5570 |
| 94.18 | Trifluormethylcarbonyl | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5419 |
| 94.19 | 2-chlorbenzoyl         | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5728 |
| 94.20 | 2-fluorbenzoyl         | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5650 |
| 94.21 | 2,4-difluorbenzoyl     | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5550 |

Table 95: Compounds of the formula



| Nr.  | R <sub>5</sub> | R <sub>6</sub> | physical data                         |
|------|----------------|----------------|---------------------------------------|
| 95.1 | ethoxycarbonyl | H              | n <sub>D</sub> <sup>20</sup> : 1.5677 |
| 95.2 | ethoxycarbonyl | methyl         | Resin                                 |

| Nr.  | R <sub>5</sub> | R <sub>6</sub> | physical data                         |
|------|----------------|----------------|---------------------------------------|
| 95.3 | ethoxycarbonyl | isopropyl      | n <sub>D</sub> <sup>20</sup> : 1.5458 |

Formulation Examples (% = percent by weight)Example F1: Emulsifiable concentrates

|  | a)   | b)   | c)   |
|--|------|------|------|
| active ingredient                                    | 25 % | 40 % | 50 % |
| calcium dodecylbenzenesulfonate                      | 5 %  | 8 %  | 6 %  |
| castor oil polyethylene glycol ether (36 mol EO)     | 5 %  | -    | -    |
| tributylphenol polyethylene glycol ether (30 mol EO) | -    | 12 % | 4 %  |
| cyclohexanone  | -    | 15 % | 20 % |
| xylene mixture                                       | 65 % | 25 % | 20 % |

Mixing finely ground active ingredient and additives gives an emulsifiable concentrate which yields emulsions of the desired concentration on dilution with water.

Example F2: Solutions

|                                   | a)   | b)   | c)   | d)   |
|-----------------------------------|------|------|------|------|
| active ingredient                 | 80 % | 10 % | 5 %  | 95 % |
| ethylene glycol monomethyl ether  | 20 % | -    | -    | -    |
| polyethylene glycol (MW 400)      | -    | 70 % | -    | -    |
| N-methylpyrrolid-2-one            | -    | 20 % | -    | -    |
| epoxidised coconut oil            | -    | -    | 1 %  | 5 %  |
| benzine (boiling range: 160-190°) | -    | -    | 94 % | -    |

Mixing finely ground active ingredient and additives gives a solution suitable for use in the form of microdrops.

Example F3: Granules

|                               | a)   | b)   | c)   | d)   |
|-------------------------------|------|------|------|------|
| active ingredient             | 5 %  | 10 % | 8 %  | 21 % |
| kaolin                        | 94 % | -    | 79 % | 54 % |
| highly dispersed silicic acid | 1 %  | -    | 13 % | 7 %  |
| attapulgite                   | -    | 90 % | -    | 18 % |

The active ingredient is dissolved in dichloromethane, the solution is sprayed onto the carrier mixture and the solvent is evaporated off *in vacuo*.

Biological ExamplesExample B1: Action against *Heliothis virescens* caterpillars

Young soybean plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound. After the spray-coating has dried, the soybean plants are populated with 10 caterpillars of *Heliothis virescens* in the first stage and placed in a plastics container. Evaluation is made 6 days later. The percentage reduction in population and the percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on the untreated plants.

The compounds of the Tables exhibit good activity against *Heliothis virescens* in this test. In particular, the compounds 1.3, 1.4, 1.6, 1.10, 93.1, 93.11, 93.16 and 93.17 are more than 80 % effective.

Example B2: Action against *Plutella xylostella* caterpillars

Young cabbage plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound. After the spray-coating has dried, the cabbage plants are populated with 10 caterpillars of *Plutella xylostella* in the third stage and placed in a plastics container. Evaluation is made 3 days later. The percentage reduction in population and the percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on the untreated plants.

The compounds of the Tables exhibit good activity against *Plutella xylostella*. In particular, the compounds 1.4 to 1.12, 1.18, 93.1, 93.11, 93.16 and 93.17 are more than 80 % effective.

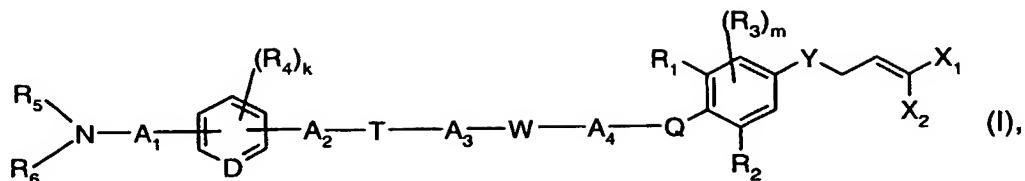
Example B3: Action against *Spodoptera littoralis*

Young soybean plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound and, after the spray-coating has dried, the plants are populated with 10 caterpillars of *Spodoptera littoralis* in the first stage and then placed in a plastics container. 3 days later, the percentage reduction in population and the percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on untreated plants.

The compounds of the Tables exhibit good activity in this test. In particular, the compounds 1.4 to 1.12, 1.18, 93.1, 93.11, 93.16 and 93.17 are more than 80 % effective.

What is claimed is

## 1. A compound of formula



wherein

$A_1$ ,  $A_2$  and  $A_3$  are each independently of the others a bond or a  $C_1$ - $C_6$ alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from  $C_3$ - $C_8$ cycloalkyl,  $C_3$ - $C_8$ cycloalkyl- $C_1$ - $C_6$ alkyl and  $C_1$ - $C_3$ haloalkyl;

$A_4$  is a  $C_1$ - $C_6$ alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from  $C_3$ - $C_8$ cycloalkyl,  $C_3$ - $C_8$ cycloalkyl- $C_1$ - $C_6$ alkyl and  $C_1$ - $C_3$ haloalkyl;

$D$  is CH or N;

$T$  is a bond, O, NH,  $NR_7$ , S, SO,  $SO_2$ ,  $-C(=O)-O-$ ,  $-O-C(=O)-$ ,  $-C(=O)-NR_8-$  or  $-NR_8-C(=O)-$ ;

$W$  is O,  $NR_7$ , S, SO,  $SO_2$ ,  $-C(=O)-O-$ ,  $-O-C(=O)-$ ,  $-C(=O)-NR_8-$  or  $-NR_8-C(=O)-$ ;

$Q$  is O,  $NR_7$ , S, SO or  $SO_2$ ;

$Y$  is O,  $NR_7$ , S, SO or  $SO_2$ ;

$X_1$  and  $X_2$  are each independently of the other fluorine, chlorine or bromine;

$R_1$ ,  $R_2$  and  $R_3$  are each independently of the others H, halogen, CN, nitro,  $C_1$ - $C_6$ alkyl,  $C_1$ - $C_6$ haloalkyl,  $C_1$ - $C_6$ alkylcarbonyl,  $C_2$ - $C_6$ alkenyl,  $C_2$ - $C_6$ haloalkenyl,  $C_2$ - $C_6$ alkynyl,  $C_1$ - $C_6$ alkoxy,  $C_1$ - $C_6$ haloalkoxy,  $C_2$ - $C_6$ alkenyloxy,  $C_2$ - $C_6$ haloalkenyloxy,  $C_2$ - $C_6$ alkynyoxy,  $C_1$ - $C_6$ alkoxycarbonyl or  $C_2$ - $C_6$ haloalkenyloxy; the substituents  $R_3$  being independent of one another when  $m$  is 2;

$R_4$  is H, halogen, CN, nitro,  $C_1$ - $C_6$ alkyl,  $C_1$ - $C_6$ haloalkyl,  $C_1$ - $C_6$ alkylcarbonyl,  $C_2$ - $C_6$ alkenyl,  $C_2$ - $C_6$ haloalkenyl,  $C_2$ - $C_6$ alkynyl,  $C_1$ - $C_6$ alkoxy,  $C_1$ - $C_6$ haloalkoxy,  $C_2$ - $C_6$ alkenyloxy,  $C_2$ - $C_6$ haloalkenyloxy,  $C_2$ - $C_6$ alkynyoxy,  $C_1$ - $C_6$ alkoxycarbonyl or  $C_2$ - $C_6$ haloalkenyloxy; the substituents  $R_4$  being independent of one another when  $k$  is greater than 1;

R<sub>5</sub> is H, CN, OH, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyloxy, -C(=O)R<sub>9</sub>, -C(=S)R<sub>9</sub>, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group consisting of halogen, C<sub>1</sub>-C<sub>6</sub>alkyl, halo-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, halo-C<sub>1</sub>-C<sub>6</sub>alkoxy, hydroxy, cyano and nitro;

R<sub>6</sub> is H, CN, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, -C(=O)R<sub>9</sub>, -C(=S)R<sub>9</sub>, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group consisting of halogen, C<sub>1</sub>-C<sub>6</sub>alkyl, halo-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, halo-C<sub>1</sub>-C<sub>6</sub>alkoxy, hydroxy, cyano and nitro; or

R<sub>5</sub> and R<sub>6</sub> together form a four- to eight-membered alkylene or a four- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>, and the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl; or

R<sub>6</sub> is -C(=O)R<sub>9</sub> or -C(=S)R<sub>9</sub>, and R<sub>5</sub> and R<sub>9</sub> together form a two- to eight-membered alkylene or a two- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>, and wherein the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl; or

R<sub>5</sub> and R<sub>6</sub> are each independently of the other -C(=O)R<sub>9</sub> or -C(=S)R<sub>9</sub>, and the two R<sub>9</sub> together form a two- to eight-membered, straight-chain or branched alkylene or a two- to eight-membered alkenylene bridge wherein a CH<sub>2</sub> group may have been replaced by O, S or NR<sub>10</sub>; and wherein the alkylene or alkenylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl, C<sub>3</sub>-C<sub>8</sub>cycloalkyl-C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, CN and -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl;

R<sub>7</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl or C<sub>3</sub>-C<sub>8</sub>cycloalkyl;

R<sub>8</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxyalkyl, -C(=O)C<sub>1</sub>-C<sub>6</sub>alkyl or C<sub>3</sub>-C<sub>8</sub>cycloalkyl;

R<sub>9</sub> is C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>2</sub>-C<sub>6</sub>alkenyloxy, C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy, C<sub>2</sub>-C<sub>6</sub>alkynyoxy, C<sub>3</sub>-C<sub>6</sub>cycloalkyl, phenyl, benzyl; or phenyl or benzyl each of which is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>6</sub>haloalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl, C<sub>2</sub>-C<sub>6</sub>alkenyl, C<sub>2</sub>-C<sub>6</sub>haloalkenyl, C<sub>2</sub>-C<sub>6</sub>alkynyl, C<sub>1</sub>-C<sub>6</sub>alkoxy, C<sub>1</sub>-C<sub>6</sub>haloalkoxy, C<sub>1</sub>-C<sub>6</sub>alkoxycarbonyl, C<sub>1</sub>-C<sub>3</sub>haloalkoxycarbonyl and C<sub>2</sub>-C<sub>6</sub>haloalkenyloxy;

R<sub>10</sub> is H, C<sub>1</sub>-C<sub>6</sub>alkyl, C<sub>1</sub>-C<sub>3</sub>haloalkyl, C<sub>1</sub>-C<sub>3</sub>haloalkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>alkoxalkyl, C<sub>1</sub>-C<sub>6</sub>alkylcarbonyl or C<sub>3</sub>-C<sub>8</sub>cycloalkyl;

k, when D is nitrogen, is 1, 2 or 3; or, when D is CH, is 1, 2, 3 or 4; and

m is 1 or 2;

and, where applicable, a possible E/Z isomer, E/Z isomeric mixture and/or tautomer thereof, in each case in free form or in salt form.

2. A compound according to claim 1 of formula (I) in free form.

3. A compound according to either claim 1 or claim 2 of formula (I) wherein X<sub>1</sub> and X<sub>2</sub> are chlorine or bromine.

4. A compound according to claim 1 of formula (I) wherein D is CH<sub>2</sub>.

5. A pesticidal composition which comprises as active ingredient at least one compound according to claim 1 of formula (I), in free form or in agrochemically acceptable salt form, and at least one adjuvant.

6. A process for the preparation of a composition as described in claim 4 which comprises intimately mixing the active ingredient with the adjuvant(s).

7. A method of controlling pests which comprises applying a pesticidal composition as described in claim 4 to the pests or to the locus thereof.

8. Use of a compound according to any one of claims 1 to 3 of formula (I), in free form or, where applicable, in agrochemically acceptable salt form, in the preparation of a composition as described in claim 4.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 03/06846

## A. CLASSIFICATION OF SUBJECT MATTER

|       |            |            |            |            |            |
|-------|------------|------------|------------|------------|------------|
| IPC 7 | C07C233/75 | C07C233/60 | C07C233/25 | C07C233/61 | C07C233/33 |
|       | C07C233/76 | C07C233/80 | C07C233/62 | C07C233/34 | C07C271/58 |
|       | C07C271/28 | C07C217/84 | A01N47/20  | A01N39/00  | A01N37/24  |

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C07C A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

CHEM ABS Data, EPO-Internal, WPI Data, PAJ, BEILSTEIN Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages   | Relevant to claim No. |
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Date of the actual completion of the international search

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## INTERNATIONAL SEARCH REPORT

International Application No

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A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 A01N33/02

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## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

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**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No

PCT/EP 03/06846

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